

**REMARKS/ARGUMENTS**

Favorable consideration of this application, as presently amended and in light of the following discussion, is respectfully requested.

Claims 1, 4-6 and 10-16 are presently pending in this application, Claims 2, 3 and 7-9 having been canceled, Claims 1 and 4-6 having been amended, and Claims 10-16 having been newly added by the present amendment.

In the outstanding Office Action, Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §101, as containing subject matters rendered inoperative and lacking utility; the specification was objected and Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §112, first paragraph, as containing subject matter not enabling to one skilled in the relevant art; Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §112, second paragraph, for being indefinite; Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §112, second paragraph, as being incomplete for omitting essential elements; Claims 1 and 4-6 were rejected under 35 U.S.C. §102(b) as being anticipated by Iwamura et al. (*Detection of Anomalous elements, x-rays, and excess heat induced by continuous diffusion of deuterium through multilayer cathode (Pd/CaO/Pd)*); Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §102(a) as being anticipated by JP 2000-042388 (hereinafter "JP '388"); and Claims 1, 2 and 4-6 were rejected under 35 U.S.C. §103(a) as being unpatentable over JP '388 in combination with JP 2000-258573 (hereinafter "JP '573").

Submitted herewith is a separate LETTER SUBMITTING DRAWING SHEET, submitting for approval changes to Figures 13 and 14. Specifically, Figures 13 and 14 have been amended to correct typographical informalities.

Applicants respectfully request that Claims 2, 3 and 7-9 be canceled without prejudice.

Newly added Claims 10-16 are believed to find clear support in the claims, specification, and drawings as originally filed. For example, Claim 10 is believed to be supported by the specification, page 15, lines 17-22, page 24, lines 19-23, page 27, lines 7-9, and page 36, lines 20-23, Claim 11 by the specification, page 18, lines 3-7, Claim 12 by the specification, page 32, lines 9-13, Claim 13 by the specification, page 16, lines 14-18, Claim 14 by the specification, page 14, lines 8-19, and Claims 15 and 16 by original Claim 1. Hence, no new matter is believed to be added thereby.

With regard to the objection to the specification and rejections under 35 U.S.C. §101 and 35 U.S.C. §112, first paragraph, Applicants respectfully traverse these rejections. In the outstanding Office Action, a large number of articles which contradict cold fusion have been cited. Most of these articles relate to the credibility of the article regarding the "Utah Experiment" which reported cold fusion for the first time in the world, and since the first article about the "Utah Experiment" did not disclose specific conditions of the experiment in detail, third parties failed in verifying the results. However, Applicants' nuclide transmutation device is believed to be completely different from that of the "Utah Experiment," and thus the citations relating to the credibility of the "Utah Experiment" are believed to be less significant in understanding the present invention.

Accordingly, Applicants respectfully invite the Examiner's attention to the references attached hereto as Exhibits A-C. These references are published after the filing date of the present application and believed to be more pertinent to the subject matters disclosed in this application. Exhibit A is *Elemental Analysis of Pd Complexes: Effects of D<sub>2</sub> Gas*

*Permeation*<sup>1</sup> (hereinafter, "Y. Iwamura et al.") written by Applicants in the present application. Y. Iwamura et al. discloses a nuclide transmutation device including a Pd complex having a Pd substrate, CaO and Pd layers and a Pd film,<sup>2</sup> and the structures of the Y. Iwamura et al. device and Pd complex are believed to be substantially the same as those disclosed as examples in Applicants' specification. Also, the preparation of a material which undergoes nuclide transmutation and the nuclide transmutation reaction of the material with D<sub>2</sub> are believed to be conducted under substantially the same conditions. For example, the Pd complex of the Y. Iwamura et al. device includes the Pd substrate with a thickness of 0.1 mm, the CaO and Pd layers with a total thickness of 1000 Å, the Pd film with a thickness of 400 Å. On such a Pd complex, a Cs layer is deposited by electrolysis using 1 mM CsNO<sub>3</sub>/D<sub>2</sub>O solution and applying a voltage of 1 V for 10 s, and then the D<sub>2</sub> gas is supplied at 1 atm in order to cause the nuclide transmutation reaction with Cs. As a result, similarly to the results described in Applicants' specification, the number of Cs atoms decreased while that of Pr atoms increased with increasing experimental time. Regarding the precision of such results, Y. Iwamura et al. states that "[t]hese results were obtained six times and reproducibility was good."<sup>3</sup> In addition, substantially the same apparatus and nuclear transmutation reaction are disclosed in *Low Energy Nuclear Transmutation In Condensed Matter Induced By D<sub>2</sub> Gas Permeation Through Pd Complexes: Correlation Between Deuterium Flux And Nuclear Products* by Iwamura et al., Tenth Int. Conf. Cold Fusion 2003 Cambridge, MA. Furthermore, a third party has performed such nuclear transmutation experiments three times, and confirmed the production of Pr.<sup>4</sup> These experimental results are believed to clearly

---

<sup>1</sup> Jpn. J. Appl. Phys. Vol. 41 (2002) pp. 4642-4650.

<sup>2</sup> Y. Iwamura et al., Figs. 2(a) and 3(a).

<sup>3</sup> Id., page 4647, right column.

<sup>4</sup> *Replication Of MHI Transmutation Experiment By D<sub>2</sub> Gas Permeation Through Pd Complex* by Higashiyama et al., Tenth Int. Conf. Cold Fusion 2003 Cambridge, MA.

support the occurrence of the nuclear transmutation reactions disclosed in the Applicants' specification. Therefore, Applicants respectfully request that the rejections under 35 U.S.C. §101 and 35 U.S.C. §112, first paragraph, be withdrawn.

With regard to the rejection under 35 U.S.C. §112, second paragraph, Claims 1 and 4-6 have been amended to clarify the subject matters recited therein. Thus, Claims 1 and 4-6 as amended are believed to be in compliance with the requirements of the statute. Also, these claim amendments are believed to find clear support in the claims, specification, and drawings as originally filed.<sup>5</sup> Furthermore, Claim 1 is amended to include subject matters recited in original Claims 2 and 3, and thus clearly supported by the original disclosure. Hence, no new matter is believed to be added by these amendments.

Briefly recapitulating, amended Claim 1 is directed to a nuclide transmutation device including a structure body including a hydrogen absorbing metal or a hydrogen absorbing alloy, an absorption part in which one surface of the structure body is exposed to a deuterium gas at a predetermined pressure, a desorption part in which another surface of the structure body is exposed to the deuterium gas at a pressure lower than the predetermined pressure in the absorption part, the desorption part and the absorption part being positioned to enclose the structure body in a closed space sealed by the structure body, a high pressurization device configured to produce the predetermined pressure in the absorption part; the high pressurization device including a deuterium supply device configured to supply the deuterium gas to the absorbing part, a low pressurization device configured to reduce the pressure in the desorption part, the low pressurization device including an exhaust gas device configured to evacuate the desorption part, and a transmutation material binding device configured to bind a material that undergoes nuclide transmutation on the one surface of the structure body. By

---

<sup>5</sup> See, for example, Specification, page 14, lines 3-7, and page 15, lines 11-16.

providing such a high pressurization device and a low pressurization device, the deuterium gas readily flows from the absorption part at a higher pressure to the desorption part at a lower pressure, thereby facilitating the absorption of deuterium in the structure body and achieving more effective nuclide transmutation reaction.<sup>6</sup>

Iwamura et al. discloses an apparatus developed to induce nuclear reactions. Nevertheless, Iwamura et al. is not believed to teach “a high pressurization device configured to produce the predetermined pressure in said absorption part, said high pressurization device including a deuterium supply device configured to supply the deuterium gas to said absorbing part” or “a low pressurization device configured to reduce the pressure in said desorption part, said low pressurization device including an exhaust gas device configured to evacuate said desorption part” as recited in amended Claim 1. On the other hand, Iwamura et al. merely discloses a continuous diffusion experimental apparatus where deuterium atoms are loaded by electrochemical potential into one side of the Pd sample and released from the other side.<sup>7</sup> Therefore, the structure recited in amended Claim 1 is believed to be clearly distinguishable from Iwamura et al.

JP '388 and JP '573 disclose a hydrogen purifying membrane and a material for in-solid nuclear reaction, respectively. However, JP '388 and JP '573 teach neither “a high pressurization device configured to produce the predetermined pressure in said absorption part, said high pressurization device including a deuterium supply device configured to supply the deuterium gas to said absorbing part” nor “a low pressurization device configured to reduce the pressure in said desorption part, said low pressurization device including an exhaust gas device configured to evacuate said desorption part” as recited in amended Claim 1. Specifically, JP '388 simply discloses a *hydrogen purifying membrane* including the Pd

---

<sup>6</sup> Specification, page 15, lines 11-16.

<sup>7</sup> Iwamura et al., Fig. 1 and page 2, lines 5-6.

substrate 16, the CaO-Pd mixture layer 17 and the Pd layer 18.<sup>8</sup> Likewise, JP '573 merely discloses the material including the Pd substrate 11, the CaO layer 5 and the outer film 8.<sup>9</sup> Therefore, the structure recited in Claim 1 is believed to be clearly distinguishable from JP '388 and JP '573.

Because none of Iwamura et al., JP '388 and JP '573 discloses the high pressurization device and the low pressurization device as recited in amended Claim 1, even the combined teachings of these cited references are not believed to render the structure recited in Claim 1 obvious.

Likewise, independent Claim 16 is believed to include subject matter substantially similar to what is recited in Claim 1 to the extent discussed above. Thus, Claim 16 is also believed to be distinguishable from Iwamura et al., JP '388 and JP '573.

For the foregoing reasons, Claims 1 and 16 are believed to be allowable. Furthermore, since Claims 4-6 and 10-15 ultimately depend from Claim 1, substantially the same arguments set forth above also apply to these dependent claims. Hence, Claims 4-6 and 10-15 are believed to be allowable as well.

---

<sup>8</sup> JP '388, Abstract and Fig. 4.

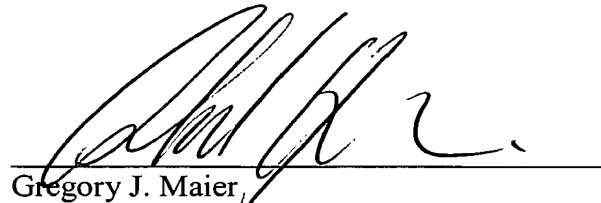
<sup>9</sup> JP '573, Abstract and Fig. 1.

Application No. 09/981,983  
Reply to Office Action of October 28, 2003

In view of the amendments and discussions presented above, Applicants respectfully submit that the present application is in condition for allowance, and an early action favorable to that effect is earnestly solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,  
MAIER & NEUSTADT, P.C.

A handwritten signature in dark ink, appearing to read 'G. Maier', is written over a horizontal line.

Gregory J. Maier  
Attorney of Record  
Registration No. 25,599

Customer Number  
**22850**

Tel: (703) 413-3000  
Fax: (703) 413 -2220  
(OSMMN 08/03)

Akihiro Yamazaki  
Registration No. 46,155

GJM/AY/YO:fm  
\\ZEUS\PACIFIC\FILES\ATTY\YO\21s\215141\AME\_MODIFIED.D  
OC

## Elemental Analysis of Pd Complexes: Effects of D<sub>2</sub> Gas Permeation

Yasuhiro IWAMURA\*, Mitsuru SAKANO and Takehiko ITOH

Advanced Technology Research Center, Mitsubishi Heavy Industries Ltd., 1-8-1 Sachiura, Kanazawa-ku, Yokohama 236-8515, Japan

(Received July 18, 2001; revised manuscript received February 1, 2002; accepted for publication April 9, 2002)

Elemental analysis of Pd complexes, which consist of a thin Pd layer, alternating CaO and Pd layers and bulk Pd, is described, after subjecting the Pd complexes to D<sub>2</sub> gas permeation. The Pd complex was located in a vacuum chamber and the elemental analysis was performed using an X-ray photoelectron spectroscopy (XPS) apparatus mounted on the chamber. When Cs was added on the surface of a Pd complex, Pr emerged on the surface while Cs decreased after the Pd complex was subjected to D<sub>2</sub> gas permeation at 343 K and 1 atm for about one week. In the case of adding Sr on the surface, Mo emerged on the surface while the added Sr decreased after D<sub>2</sub> permeation for about two weeks. All the phenomena were reproduced qualitatively. The isotopic composition of the detected Mo exhibited characteristics indicating an isotopic abundance of Sr rather than the natural abundance of Mo. [DOI: 10.1143/JJAP.41.4642]

**KEYWORDS:** D<sub>2</sub> gas, permeation, Pd, Pd complex, thin film, Cs, Pr, Sr, Mo, isotopic composition, X-ray photoelectron spectroscopy

### 1. Introduction

The palladium–hydrogen system has been the most widely investigated system among the metal hydrides since it has the following features: (1) Pd exhibits high solubility and diffusivity of hydrogen; Pd and Pd alloys have been used as purification membranes of hydrogen.<sup>1)</sup> (2) Pd hydride is a super-conducting material<sup>2)</sup> that attracted the attention of researchers in the 1970s–80s. However, interest in Pd hydride as a practical super-conducting material has declined because other materials that show higher transition temperatures have been discovered. (3) In the palladium–deuterium system, several researchers, mainly in recent years, have claimed low-energy transmutations,<sup>3–8)</sup> although a definite theoretical explanation has not been provided.

We have been investigating the low-energy transmutations observed in the palladium–deuterium system. Our experimental results thus far have led to the assumption that necessary conditions which give rise to the phenomena should satisfy the following criteria:<sup>7)</sup> (i) existence of a low work function material, typically CaO, near the Pd surface; (ii) sufficient diffusion flux of deuterium, (iii) sufficient D on the Pd surface. To check the validity of these assumptions, we have established an experimental method characterized by “permeation of D through Pd complex” as shown in Fig. 1. D<sub>2</sub> gas molecules cause a dissociative chemisorption on the D<sub>2</sub> gas side of the Pd complex, and then D atoms diffuse toward the vacuum side, where they combine and are released as D<sub>2</sub> gas. We assume that the above necessary conditions are satisfied on the near surface of D<sub>2</sub> gas side of the Pd complex.

The Pd complex is composed of a Pd thin film, alternating CaO and Pd layers and bulk Pd as shown in Fig. 2(a). This composition was determined based on condition (i) above since the work function of CaO is relatively low (1.6 eV–1.86 eV).<sup>9)</sup> Permeation of deuterium through the Pd complex is attained by exposing one side of the Pd complex to D<sub>2</sub> gas while maintaining the other side under vacuum conditions. We assume that D<sub>2</sub> gas permeation satisfies the conditions (ii) and (iii) above. Although the definite physical implica-

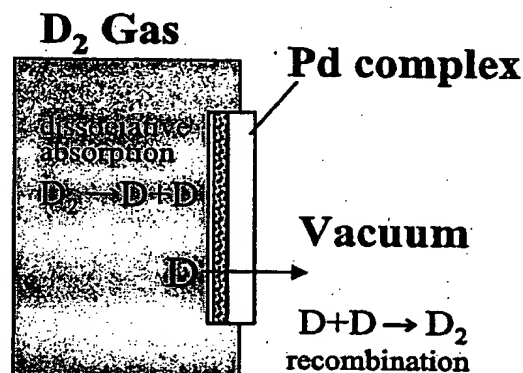


Fig. 1. D<sub>2</sub> gas permeation through the Pd complex.

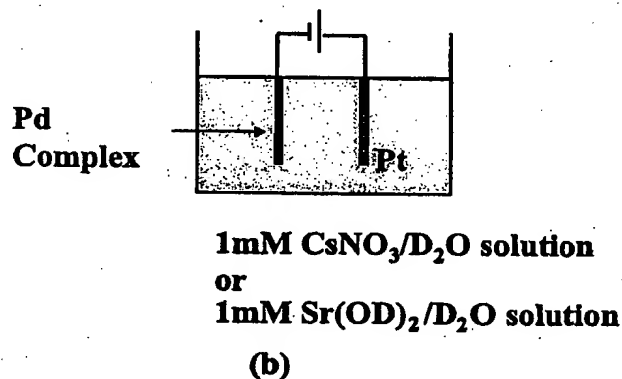
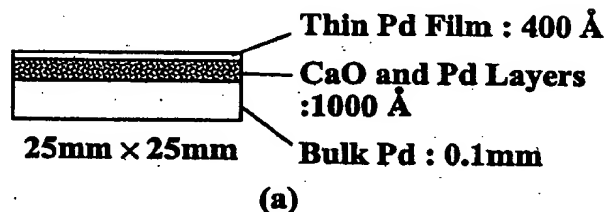


Fig. 2. (a) Structure of Pd complex, (b) Method of depositing Cs or Sr on the surface of Pd complex.

\*Present address: Takasago Research and Development Center, Mitsubishi Heavy Industries Ltd., 2-1-1 Shinhaman Arai-cho, Takasago, Hyogo 676-8686, Japan.



tions of the above conditions are unclear at present; it is a fact that the experimental observations described in this paper are consistent with the assumptions.

Almost all the other researchers in this field are conducting experiments involving the electrolysis of  $D_2O$  or  $H_2O$  with metal as a cathode<sup>3-5)</sup> and they usually analyze the transmuted products on the cathode metal by taking it out of the experimental apparatus after electrolysis. Our experimental method described above is superior in that it discriminates the products from contamination because we analyze the products in vacuum during the experiments without moving the products.

## 2. Experimental

Figure 2(a) shows a schematic view of the Pd complex cross section. The Pd complex is composed of a Pd thin film, alternating CaO and Pd layers and bulk Pd. Test pieces were prepared by the following procedures. A Pd plate (with purity up to 99.99%; Tanaka Kikinzoku Kogyo K.K.) was washed with acetone and annealed in vacuum ( $<10^{-7}$  Torr) at 900°C for 10 h. It was then cooled to room temperature in furnace and washed with D-based aqua regia (with purity up to 99.9%), which consisted of a mixed liquid of  $DCl/D_2O$  and  $DNO_3/D_2O$ , to remove impurities on the surface of the Pd plate. The surface of the plate was covered by layers of CaO and Pd (1000 Å) which were obtained by five times alternately sputtering 20-Å-thick CaO and 180-Å-thick Pd layers. The thicknesses of the layers were measured by a crystal thickness monitor. Then a 400-Å-thick Pd layer was sputtered on the surface of the CaO and Pd layers. The thickness of a 400 Å is chosen because it is sufficient to form a bulk layer. These processes are performed by Ar ion beam sputtering, which is a process whereby material is sputtered from a target with an Ar ion beam and deposited onto a substrate. The samples are sputtered at room temperature and the voltage and current of the Ar beam are 1 kV and 20 mA, respectively. Films deposited by ion beam sputtering exhibit the property of low contamination due to the low background pressure that leads to reduced gas incorporation.

After forming a Pd complex, Cs or Sr was deposited on the surface of the thin Pd layer. Cs or Sr atom was deposited by applying an electric field to 1 mM  $CsNO_3$  (with purity up to 99.9%) or  $Sr(OD)_2$  (with purity up to 99%) solution as shown in Fig. 2(b). A Pt wire (with purity up to 99.9%) was utilized as a counter-electrode. A 1 V negative voltage was applied to the Pd complex for 10 s. The value of 1 V was chosen to suppress  $D_2$  and  $O_2$  gas generations by electrolyzing the  $CsNO_3$  or  $Sr(OD)_2$  solution.

Figure 3(a) shows a schematic of the experimental apparatus. One advantage of this newly developed apparatus is that we can analyze the surface of a Pd complex sample by X-ray photoelectron spectroscopy (XPS) without removing it from the vacuum chamber, thereby preventing contamination of the test sample. XPS is a surface analysis technique which is used to investigate the chemical composition of surfaces by irradiating a sample with monoenergetic soft X-rays and analyzing the energy of the photoelectrons. Another advantage is that we can detect the time dependence of the elements on the Pd complex surface. Repeating  $D_2$  gas permeation and XPS analyses, we can obtain information on time variation of the detected elements.

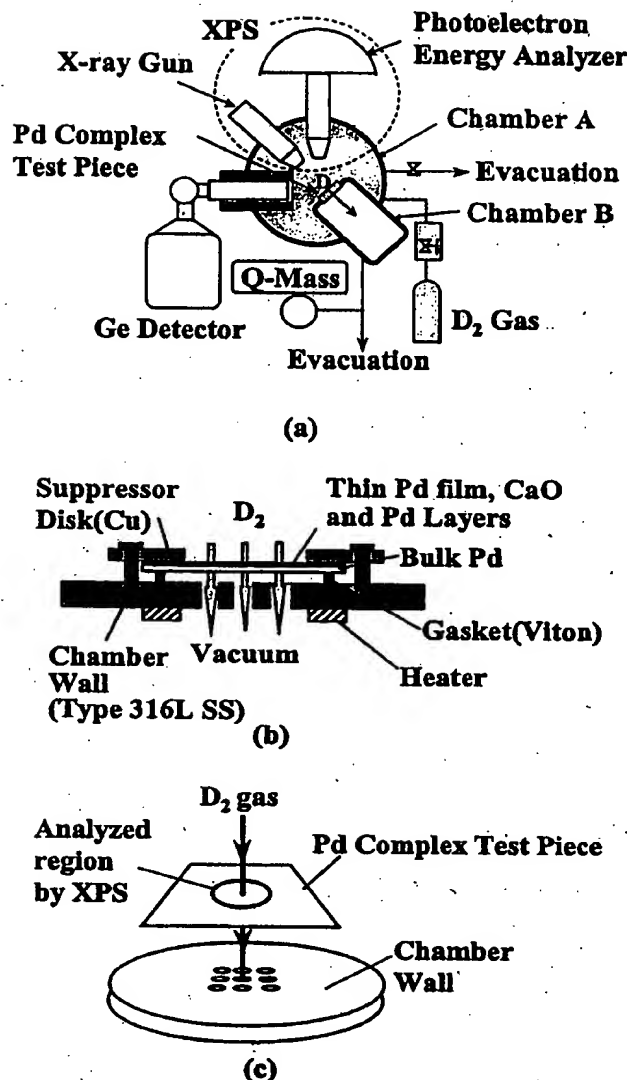


Fig. 3. (a) Experimental apparatus, (b) Schematic of test setup in the vicinity of Pd complex test piece, (c) Path of  $D_2$  gas flowing through Pd complex test piece and chamber wall.

The apparatus consists of two chambers, A and B, an X-ray gun, an electrostatic analyzer for XPS, a mass spectrometer and a Ge semiconductor detector. The mass spectrometer is installed for analyzing the  $D_2$  gas permeating the Pd complex. The Ge semiconductor detector is mounted for the detection of gamma rays. The X-ray gun is non-monochromatic and  $Al-K \alpha$  is utilized. The power is 200 W, voltage is 10 kV and current is 20 mA. The electrostatic analyzer is equipped with a five-stage electrostatic lens and the pass energy is 100 eV. The X-ray gun and the electrostatic analyzer were made in our laboratory and the energy was calibrated using Au.

One chamber (Chamber A) is filled with  $D_2$  gas, and the other chamber (Chamber B) is evacuated by a turbomolecular pump. These two chambers are divided by a Pd complex test piece.  $D_2$  gas is supplied at 1 atm on the Pd film side of the test piece and dissolves in D atoms at the surface.

The D atoms intrude into the Pd thin film and diffuse through the Pd complex and then reach the surface of the bulk side, where they combine and are released as D<sub>2</sub> molecules.

Figures 3(b) and 3(c) illustrate magnified views of an installed test piece and D<sub>2</sub> gas flow. The thin Pd film (400 Å) side of the Pd complex test piece is exposed to D<sub>2</sub> gas and the test piece is fixed by a suppressor disk, a gasket (Viton) and a chamber wall made of Type 316L SS as shown in Fig. 3(b). The stainless steel chamber wall has several small holes that connect the D<sub>2</sub> gas side with the vacuum side. D<sub>2</sub> gas passes through the Pd complex test piece as shown in the Figs. 3(b) and 3(c). The total surface area of the holes is 1.0 cm<sup>2</sup>. A heater is installed under the chamber wall and maintains the Pd temperature.

XPS analyses are performed with the following assumptions: 1) Intensities of incoming X-rays and the irradiated region are constant. 2) The analyzed region is a circle 5 mm in diameter and 20 Å in depth. The depth corresponds to the average path length of photoelectrons. 3) Atomic concentration of a detected element is calculated by the ratio of Pd peak intensity and the ionizing cross section of the element. The cross section at 1486 eV<sup>10)</sup> is used because the target of the X-ray gun is Al.

The experimental procedure is as follows. The vacuum chambers are baked sufficiently before the experiments. A Pd complex test piece with added Cs or Sr is mounted on a flange in an air environment and then introduced into the vacuum chamber. Step 1: Both of the vacuum chambers are evacuated by a turbo-molecular pump and the test piece is heated to 70°C. The ultimate pressure in the vacuum chamber was about  $3 \times 10^{-8}$  Torr. Step 2: The surface of the test piece is analyzed by XPS to confirm that no other elements on the surface of the Pd test piece are detected except the given element (Cs or Sr), Pd and C. As is widely recognized in the field of surface science, a small amount of C is usually detected on a metal surface even in the vacuum chamber. Step 3: Chamber A is filled with D<sub>2</sub> gas up to 1 atm so that deuterium atoms permeate from this side to Chamber B behind the test piece. Step 4: After a certain period (from two days to one week) of deuterium diffusion through the Pd complex test piece, the D<sub>2</sub> is evacuated from Chamber A and the surface of the Pd test piece is analyzed by XPS in the chamber. New elements that did not exist on the test piece at the beginning of the experiment can be detected. Usually this process is repeated a few times to observe the time dependence of the given or newly generated elements. Step 5: The heater is turned off and all the chambers are filled with 1 atm N<sub>2</sub> gas. The test piece is removed from the chamber and its surface is analyzed by secondary ion mass spectrometry (SIMS). SIMS is a surface analysis technique in which energetic primary ions impact the surface and generate secondary ions, which are subsequently mass-separated and detected. SIMS is capable of analyzing all the elements with isotopic discrimination. In this study, we analyzed the Pd complex test piece after an experiment using SIMS to investigate the isotopic compositions of the detected elements. Only surface isotopic compositions were analyzed; we did not measure their depth profiles. SIMS analysis was performed by Probion Analysis, Inc. in France using the Physical Electronics 6600.

### 3. Results

Experimental results are shown in Figs. 4–9. During these experiments, we did not observe statistically significant gamma ray emission and gaseous products exceeding the detection limits (about 1 ppm) of the mass spectrometer.

Experimental results obtained for test pieces deposited with Cs are shown in Fig. 4. Figure 4(a) shows the time dependence of the number of Cs and Pr atoms. Results for two runs are shown as examples. No Pr was detected at the beginning of the experiments. The number of Cs atoms decreased while that of Pr atoms increased with increasing experimental time. The amount of deuterium permeation was proportional to the elapsed time. At 120 h, the number of Pr atoms exceeded that of Cs atoms as shown in Fig. 4(a). Similar results were obtained in four other runs, although the initial number of Cs atoms was smaller than that in the Fig. 4(a). The reproducibility of these experiments is thought to be good.

The time variations in XPS spectra for the first run are shown in Figs. 4(b)–4(d) for Cs, Pr and Pd, respectively. All the elements are identified by two binding energy peaks<sup>11)</sup> of 3d<sub>5/2</sub> and 3d<sub>3/2</sub>. In Fig. 4(c), the peak ratio of 3d<sub>5/2</sub> to 3d<sub>3/2</sub> should be 3 : 2,<sup>11)</sup> however, it seems the two peaks are almost the same. The atomic ratio of Pr to Pd is a few percent and the major element is Pd. Therefore background signals derived from the photoelectrons of Pd–3d or 3p seem to influence the peak ratio of Pr. The error for Pr 3d<sub>5/2</sub> and 3d<sub>3/2</sub> is supposed to be a few cps. A peak between Pr 3d<sub>5/2</sub> and 3d<sub>3/2</sub> can be recognized around 939 eV. This is assumed to be derived from a shake-up or shake-off plasmon. It is evident that the two Cs peaks decrease corresponding to the increase in Pr peaks. On the other hand, XPS peaks for Pd do not change. This demonstrates that there is no problem with the stability of the obtained XPS spectra. Figure 4(e) shows a wide-range spectrum for the first run. Only Pd and C can be seen on the wide spectrum because the atomic ratios of Cs and Pr are a few percent. As mentioned earlier, C is usually detected on the metal surface even in the vacuum chamber.

In contrast, bulk Pd and the thin Pd film test piece with Cs deposition, which does not belong to a Pd complex, do not exhibit anomalous phenomena. All experimental conditions were the same as those of the Pd complex (Pd/CaO/Pd) experiments except for the formation of CaO and Pd layers. The annealing, etching by aqua regia, formation of a 400-Å-thick Pd thin film and deposition of Cs were performed in the same manner. Time variation of the Cs atom number, XPS spectra for Cs and Pr are given in Fig. 5. It is clear Cs did not change and Pr never appeared. This is quite reasonable in the sense that D<sub>2</sub> gas permeation usually does not affect the composition of the elements on the Pd surface. This experiment indicates that a CaO and Pd complex layer is essential for observing the decrease in Cs and the increase in Pr.

As a comparative study, we used H<sub>2</sub> gas instead of D<sub>2</sub> gas. The experimental results are shown in Fig. 6. In this case, only the gas is different; the other experimental conditions are all the same as Fig. 4. We also find that Cs did not change and Pr never appeared. This suggests that deuterium is a necessary factor for observing the anomalous behavior of Cs and Pr.

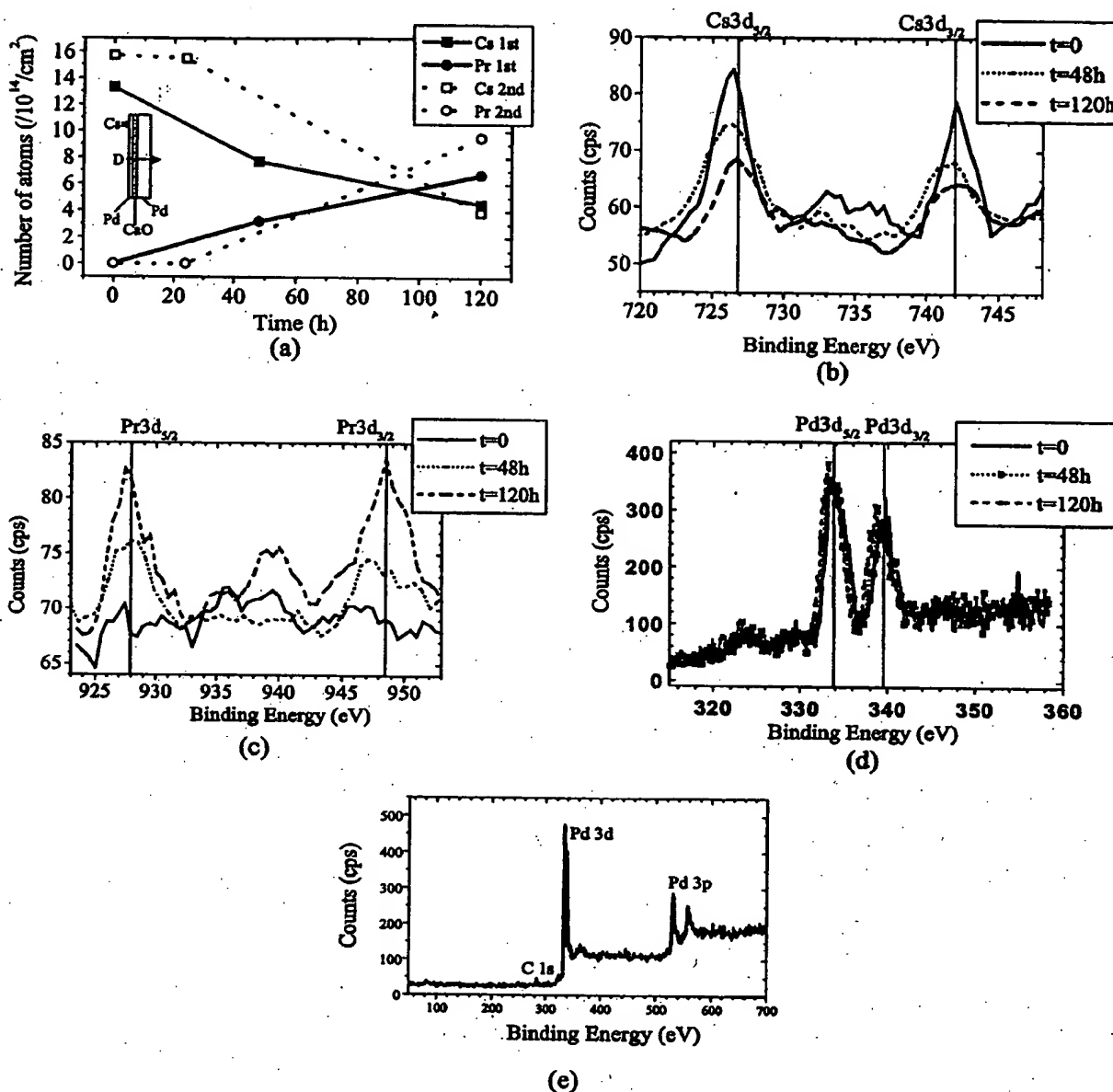


Fig. 4. Experimental results obtained by  $\text{D}_2$  gas permeation through Pd complex (Pd/CaO/Pd) deposited with Cs: (a) Time variation in number of Cs and Pr atoms (number of atoms per  $\text{cm}^2$ ), (b) XPS spectrum of Cs for experiment run #1, (c) XPS spectrum of Pr for experiment run #1, (d) XPS spectrum of Pd for experiment run #1, (e) Wide-range XPS spectrum for experiment run #1.

The experimental results for Pd complex test pieces with added Sr are shown in Fig. 7. We observed that Sr decreased while Mo increased with increasing experimental time. Figure 7(a) shows the time variation of the number of Sr and Mo atoms. Experiments were performed three times and all the data are plotted in Fig. 7. At the beginning of the experiments, no Mo atoms were detected, however, Mo atoms increased gradually while Sr decreased correspondingly. It should be noted that runs for Sr are required to be longer to convert Sr to Mo than in the case of Cs to Pr. As can be seen in Fig. 7, qualitative reproducibility is good. The time variations of XPS spectra for the first run are shown in Figs. 7(b)–7(d) for Sr, Mo and Pd, respectively. All

the elements are identified by two binding energy peaks<sup>11)</sup> in a similar way as for the Cs experiments. It is clear that the Sr peaks decrease corresponding to the increase in Mo peaks. The peaks of Pd did not change as shown in Fig. 7.

Figure 8 shows the results of the test piece devoid of CaO and Pd layers. All conditions of this comparative experiment were the same as those of the Pd complex (Pd/CaO/Pd) experiments except for the formation of CaO and Pd layers. We can see that Sr did not change and Mo was not detected. If we perform an experiment with a test sample devoid of CaO, we cannot observe any changes of elements on the surface of the sample. The CaO and Pd layers are considered to be an important factor for observing the decrease of Sr

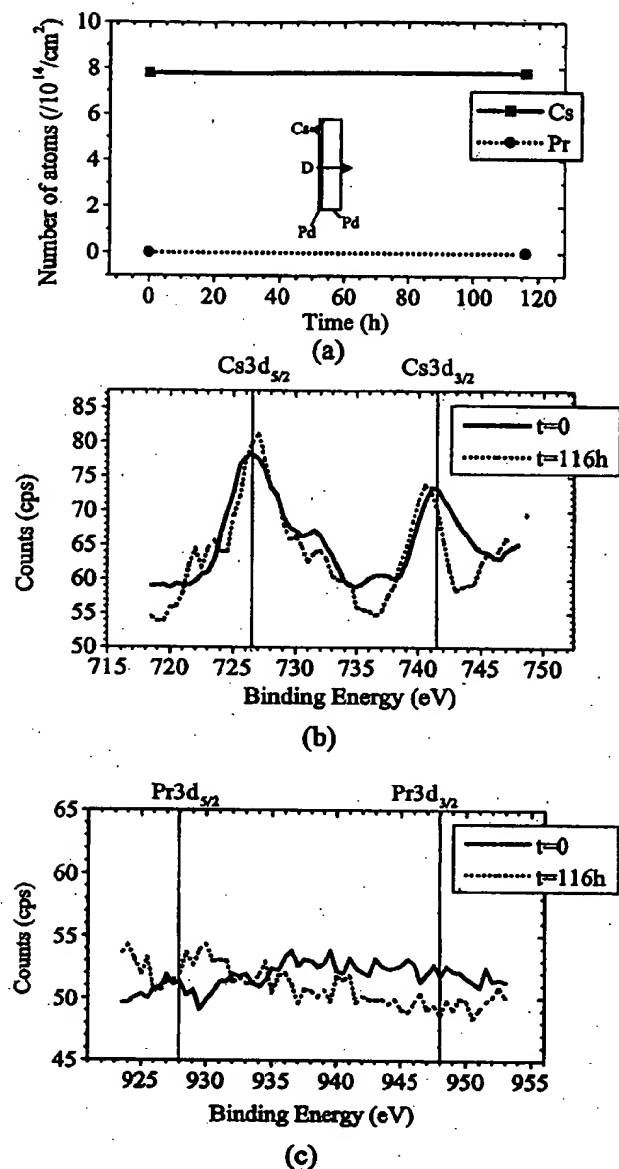


Fig. 5. Experimental results obtained by  $\text{D}_2$  gas permeation through thin film and bulk Pd with added Cs: (a) Time variation in number of Cs and Pr atoms, (b) XPS spectrum of Cs, (c) XPS spectrum of Pr.

and the increase of Mo; this is the same as the case of Cs.

The authors performed a comparative experiment using  $\text{H}_2$  gas for a Pd complex test piece with added Sr. The elements on the surface of the Pd complex test piece did not increase or decrease as expected. Since all the other experimental conditions except  $\text{H}_2$  gas are the same as those of Fig. 7, it is suggested that deuterium is a necessary factor for observing the decrease in Sr and increase in Mo.

In order to investigate the isotopic composition, the samples after the experiment were analyzed by SIMS. Figures 9(a)–9(c) show the results of SIMS analysis for the three samples described in Fig. 7. The primary ions of the SIMS analysis were  $\text{O}_2^+$  with the energy of 5.5 keV and the intensity of 30 nA. The area analyzed by SIMS was  $150\text{ }\mu\text{m}$

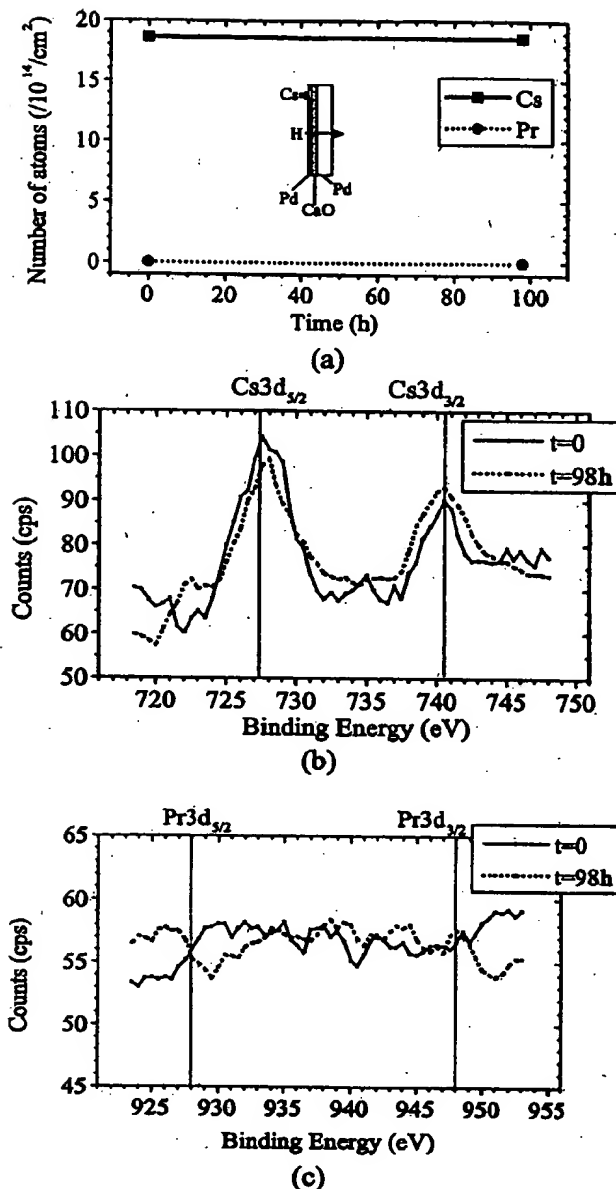


Fig. 6. Experimental results obtained by  $\text{H}_2$  gas permeation through Pd complex (Pd/CaO/Pd) with added Cs: (a) Time variation in number of Cs and Pr atoms, (b) XPS spectrum of Cs, (c) XPS spectrum of Pr.

in diameter and mass resolution was 300. The intensities of mass number 96 were the largest for each sample, although the intensities were different. Figure 9(d) shows the results of SIMS analysis for a test piece with added Sr, however, no  $\text{D}_2$  gas permeated through it. Signals arising from Mo impurity and organic compounds in Pd can be seen. The SIMS mass spectrum for a Mo layer (400 Å thickness) which was deposited on a Pd disk is shown in Fig. 9(e). This spectrum reveals the natural abundance of Mo. Comparing these figures, we can easily recognize that the isotopic compositions of the detected Mo in Figs. 9(a)–9(c) are different from the natural isotopic abundance of Mo.

The difference in the ion intensities of the three samples is

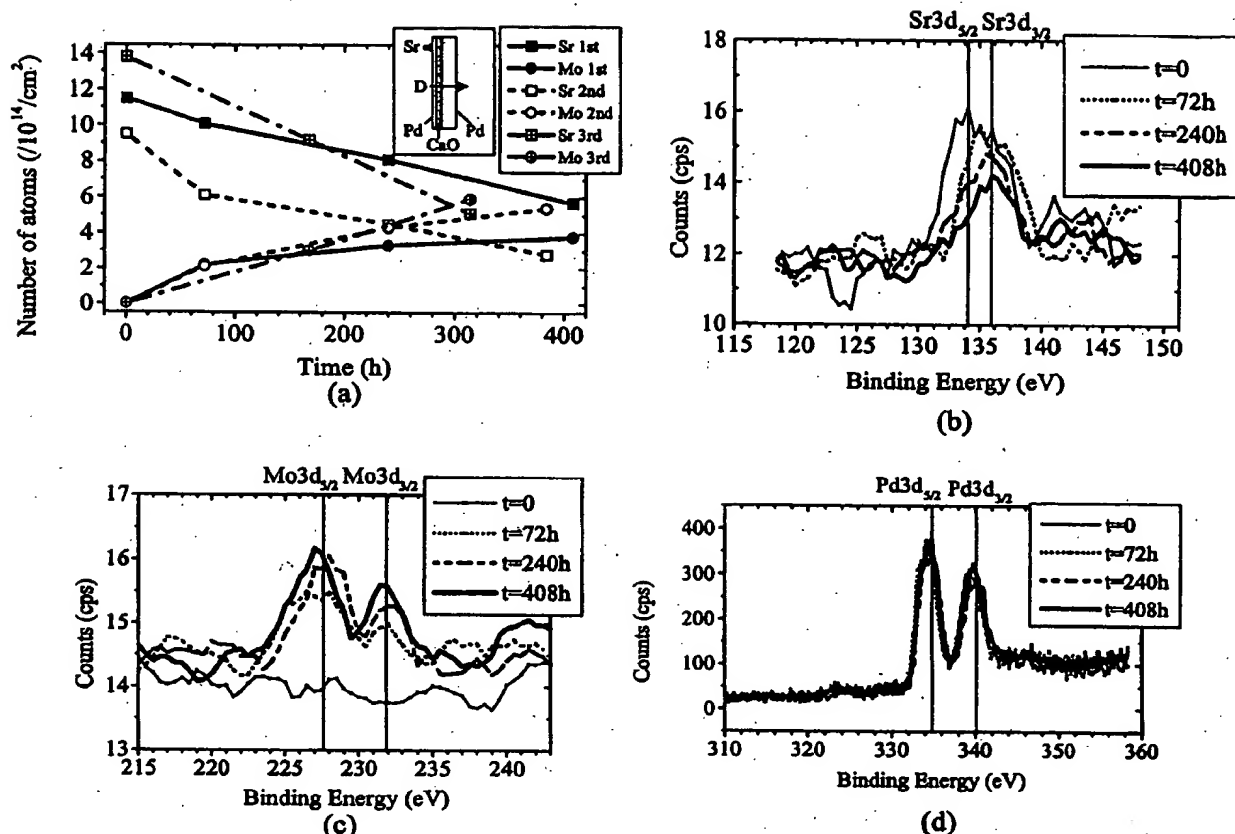


Fig. 7. Experimental results obtained by  $\text{D}_2$  gas permeation through Pd complex (Pd/CaO/Pd) with added Sr: (a) Time variation in number of Sr and Mo atoms (number of atoms per  $\text{cm}^2$ ), (b) XPS spectrum of Sr for experiment run #1, (c) XPS spectrum of Mo for experiment run #1, (d) XPS spectrum of Pd for experiment run #1.

presumed to exist that Sr and Mo did not distribute uniformly. The areas analyzed by SIMS and XPS are  $150\ \mu\text{m}$  and  $5\ \text{mm}$  in diameter, respectively. Therefore the signal analyzed by XPS is averaged over a large area, however the signal analyzed by SIMS is sensitive to the location of the analysis. If the distribution of Mo is not uniform, the result of SIMS analysis depends on the analyzed location. As we did not perform depth analysis of Mo, the dependence of isotopic compositions is not clear.

The SIMS analyses were performed by the "offset voltage technique"<sup>12)</sup> to suppress the effects of molecular ions on mass spectra. The offset voltage technique is based on the fact that the kinetic energy distribution of atomic and molecular ions is entirely different and the distribution of atomic ion energy is broader than that of molecular ions. If we filter the central part of the distribution by applying a certain offset voltage, all signals decrease, however, the signal of molecular ions decreases much more than that of atomic ions. Therefore we can select only the signals of atomic ions. In our case, an 80 V offset voltage was applied. As for the formation of molecular ions, we should consider D, H, C, O, Pd and Sr; however, it is very difficult to form a mass number 96 by combining these elements. Moreover, Mo atoms were detected by XPS; it is natural to consider that almost all of the signal of mass number 96 can be

attributed to  $^{96}\text{Mo}$ .

#### 4. Discussion

The experimental results shown in Figs. 4–6 are summarized as follows: Cs atoms decrease and, correspondingly, Pr atoms emerge on the surface of the Pd complex by permeation of  $\text{D}_2$  gas through it. These results were obtained six times and reproducibility was good. However, when we eliminate CaO and Pd complex layers from the Pd complex, or when we let  $\text{H}_2$  gas permeate through the Pd complex, we observed no decrease in Cs and no emergence of Pr. Is it possible to interpret the phenomenon based on the established chemical or physical processes? The authors do not recall any processes other than nuclear transmutation; of course, there should be careful consideration of the possible contamination of test pieces and  $\text{D}_2$  gas from the environment. The contamination issue will be discussed later in this paper. If the observed phenomenon is associated with nuclear transmutation, the following expression can be derived, as Cs and Pr each have only one isotope.



It should be noted that the mass number and the atomic number increased by 8 and 4, respectively; in this expression. It is natural to consider that deuterium plays an

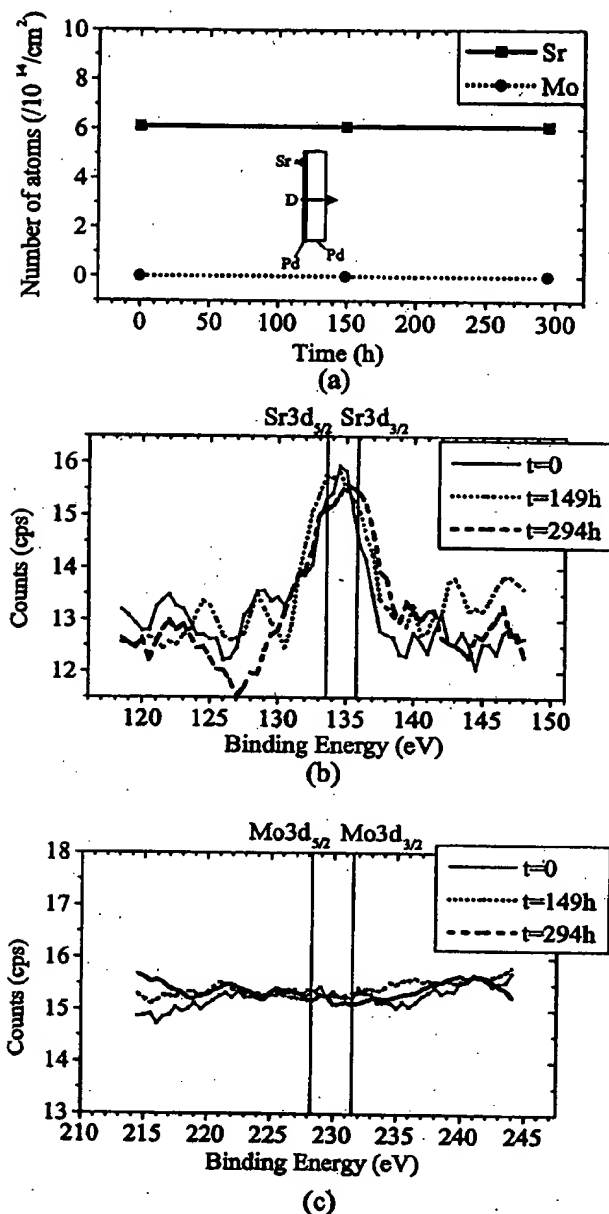


Fig. 8. Experimental results obtained by  $\text{D}_2$  gas permeation through thin film and bulk Pd deposited with Sr: (a) Time variation in number of Sr and Mo atoms, (b) XPS spectrum of Sr, (c) XPS spectrum of Mo.

important role in this phenomenon. This is due to the fact that Cs atoms decrease and, correspondingly, Pr atoms emerge on the surface of the Pd complex as  $\text{D}_2$  gas permeates through it.

The released energy for eq. (1) can be estimated by mass defects if we postulate that four deuterons react with Cs. About 50.5 MeV should be released per reaction. Figure 4(a) shows that there exist about  $10^{14}$  atoms of Pr at the surface of the test piece because the reaction surface area is  $1.0\text{ cm}^2$ . Therefore released energy can be estimated to be 800 J. The average power is calculated to be 1.8 mW when the reaction time is assumed to be 120 h. Even if 1.8 mW were released

as heat during the experiment, we could not detect or notice the released heat.

We now discuss the Sr experiments. As indicated in Fig. 7, more permeating time is necessary to convert Sr into Mo than Cs experiments. In other words, Cs is easier to change than Sr. It might be presumed that the cross sections in this low-energy transmutation process depend on the type of nuclei. If we postulate the existence of this low-energy transmutation process it might be predictable. The observed results are expressed by the following expression because Sr and Mo have several isotopes.



Figures 10(a) and 10(b) show the relationship of mass numbers between given Sr and detected Mo. The characteristic feature of the detected Mo is that mass number 96 is the largest of the isotopes of Mo for all three cases of the Sr experiments. On the other hand, the major isotope of Sr is mass number 88. This fact suggests that the following expression can explain the results of SIMS analysis.



It is also noted that the mass number and the atomic number increased by 8 and 4, respectively, in this expression, as is the case with Cs. The released energy for eq. (3) is estimated to be 53.5 MeV per reaction. Therefore we cannot detect the released heat during experiments, as is the case with Cs.

Let us discuss the contamination issue. The first point is that the XPS analysis of the nuclear products is performed in the vacuum chamber. We do not expose the Pd complex test pieces to an air environment during experiments, and we monitor pressure in the chamber. Monitoring pressure enables us to detect any gaseous contaminants invading into the chamber, as the test apparatus is located in a clean room where temperature and humidity are kept constant.

The second point is that since the detected material, Pr, belongs to rare earth elements, it is difficult to assume that Pr accumulated on the Pd complex test pieces by an ordinary process. The purity of the used  $\text{D}_2$  gas is over 99.6% and most of the impurity is  $\text{H}_2$ . The other impurities detected by a mass spectrometer are  $\text{N}_2$ ,  $\text{D}_2\text{O}$ ,  $\text{O}_2$ ,  $\text{CO}_2$ , CO and hydrocarbons; their amounts are all under 10 ppm. We analyzed Pd complex test pieces deposited with Cs by glow discharge mass spectrometry and confirmed that the amount of Pr in the test pieces was under the detection limit (0.01 ppm). If we assume that all of the Pr at 0.01 ppm distributed in the Pd test piece (0.1 mm, 0.7 g) gathered in the analyzed area, a circle of 5 mm diameter and 20 Å depth from the surface of the test piece, the number of Pr atoms attained the same order ( $10^{13}$  atoms) of the detected Pr. However, it is impossible for all of the distributed Pr in the Pd test piece to gather in the narrow surface area against the flow of  $\text{D}_2$  gas without the application of a specific force on Pr, because such a phenomenon breaks the law of thermodynamics.

The third point is that the isotope ratio of produced elements is anomalous. In this paper, we show the isotopic anomaly of Mo. It provides evidence that the detected material, Mo, was produced by certain nuclear processes. If the Mo were a contaminant, such efficient isotope separa-

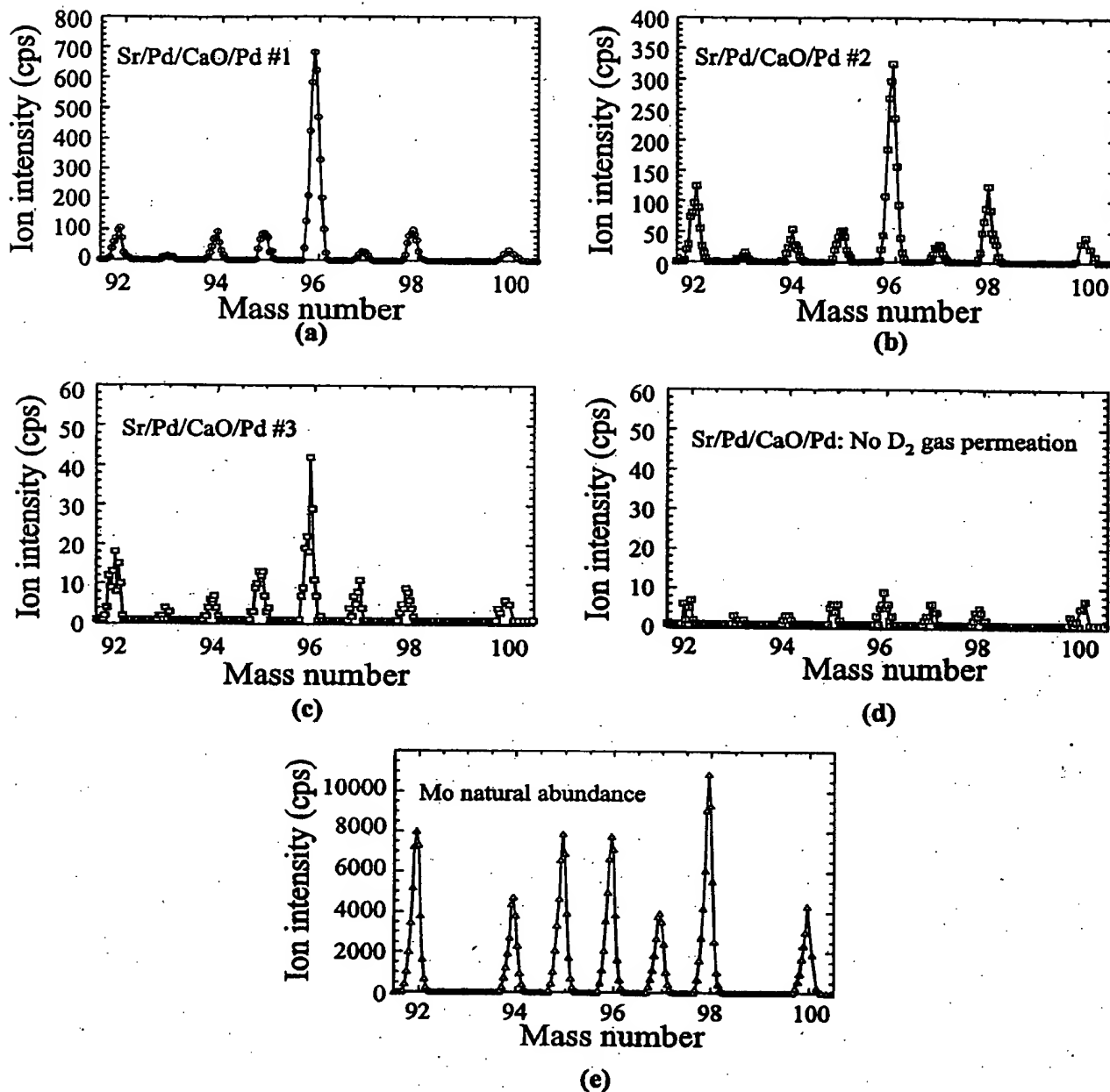


Fig. 9. Anomalous isotopic composition of detected Mo: (a) Isotopic composition of detected Mo for run #1, (b) Isotopic composition of detected Mo for run #2, (c) Isotopic composition of detected Mo for run #3, (d) SIMS analysis for Pd complex test piece with added Sr without D<sub>2</sub> gas permeation, (e) Natural abundance of Mo analyzed by SIMS.

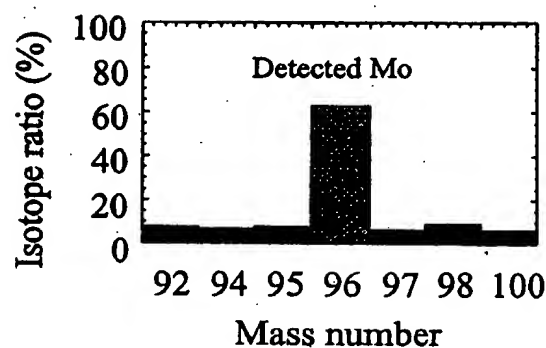
tions would not be possible.

The last point is that the elements detected by the D<sub>2</sub> gas permeation vary depending on the given elements at the beginning of the experiments. In our experiments, Pr or Mo was detected when Cs or Sr was deposited on the surface of Pd complex test pieces, respectively. It is very difficult to assume that the detected elements change depending on the given elements by external contamination.

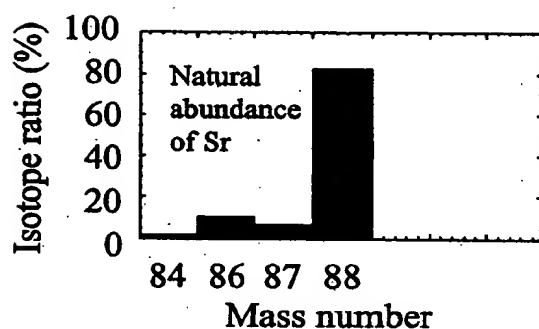
The above discussion strongly suggests the existence of low-energy nuclear transmutations induced by a simple method. We should stress, however, that impurity control of

Pd test pieces and the apparatus is an important factor to observe the transmutation process. In other words, we might not observe it if we perform experiments in a dirty environment.

We noticed that a certain rule exists between given and produced elements.<sup>13)</sup> The increase in mass number is 8, and the increase in atomic number is 4. At present, we do not have a complete theory that can explain the obtained experimental results without a few assumptions. However, if several assumptions are accepted, they are basically explained by the EINR model,<sup>5)</sup> which is one of the working



(a)



(b)

Fig. 10. Relationship of mass numbers between given Sr and detected Mo: (a) Isotopic composition of detected Mo, (b) Isotopic composition of given Sr.

hypotheses in the investigation of the nature of this phenomenon. It might be said that the observed transmutation processes belong to the new category of nuclear reactions on the metal surface associated with deuterium and electrons under low-energy conditions. However, there are many issues to be resolved, especially theoretical ones, and further investigations by other researchers are necessary.

#### Acknowledgements

The authors would like to thank N. Murakami, T. Wada, S. Kuribayashi, I. Toyoda, W. Kawamura, N. Gotoh, T. Sakai, K. Yoshikawa, Y. Tanaka, T. Kawakita and F. Kudough for valuable discussions.

- 1) F. A. Lewis: *The Palladium Hydrogen System* (Academic Press, London, 1967).
- 2) J. P. Burger: *J. Less-Common Met.* 101 (1984) 53.
- 3) T. Mizuno, T. Ohmori and M. Enyo: *Electrochemistry* 64 (1996) 1160.
- 4) T. Ohmori, T. Mizuno and Y. Nodasaka: *Fusion Technol.* 33 (1998) 367.
- 5) Y. Iwamura, T. Itoh, N. Gotoh and I. Toyoda: *Fusion Technol.* 33 (1998) 476.
- 6) G. H. Miley, G. Name, M. J. Williams, J. A. Patterson, J. Nix, D. Cravens and H. Hora: *Proc. 6th Int. Conf. Cold Fusion* (New Energy and Industrial Technology Development Organization, Tokyo, Japan, 1996) p. 629.
- 7) Y. Iwamura, T. Itoh, N. Gotoh, M. Sakano, I. Toyoda and H. Sakata: *Proc. 7th Int. Conf. Cold Fusion* (ENECO, Salt Lake, U.S.A., 1998) p. 167.
- 8) X. Z. Li, Y. J. Yan, J. Tian, M. Y. Mei, Y. Deng, W. Z. Yu, G. Y. Tang, D. X. Cao and A. De Ninno: *Proc. 8th Int. Conf. Cold Fusion* (Italian Physical Society, Bologna, Italy, 2000) p. 123.
- 9) Chemical Society of Japan: *Kagaku Binran Kisohe Kaitai* (Maruzen, Tokyo, 1984) 3rd ed. [in Japanese].
- 10) J. H. Scofield: *J. Electron Spectrosc. Relat. Phenom.* 8 (1976) 129.
- 11) M. Cardona and L. Ley: *L. Photoemission in Solid I—General Principles* (Springer-Verlag, Berlin, 1978) *Topics in Applied Physics*, Vol. 26.
- 12) R. G. Wilson, F. A. Stevie and C. W. Magee: *Secondary Ion Mass Spectrometry* (John Wiley, New York, 1987).
- 13) Y. Iwamura, T. Itoh and M. Sakano: *Proc. 8th Int. Conf. Cold Fusion* (Italian Physical Society, Bologna, Italy, 2000) p. 141.



Iwamura, Y., et al. *Low Energy Nuclear Transmutation In Condensed Matter Induced By D<sub>2</sub> Gas Permeation Through Pd Complexes: Correlation Between Deuterium Flux And Nuclear Products*. in *Tenth International Conference on Cold Fusion*. 2003. Cambridge, MA: LENR-CANR.org. This paper was presented at the 10th International Conference on Cold Fusion. It may be different from the version published by World Scientific, Inc (2003) in the official Proceedings of the conference.

## Low Energy Nuclear Transmutation In Condensed Matter Induced By D<sub>2</sub> Gas Permeation Through Pd Complexes: Correlation Between Deuterium Flux And Nuclear Products

Y. IWAMURA, T. ITOH, M. SAKANO, S. SAKAI, S. KURIBAYASHI

*Advanced Technology Research Center, Mitsubishi Heavy Industries, Ltd.  
1-8-1, Sachiura, Kanazawa-ku, Yokohama, 236-8515, Japan  
iwamura@atrc.mhi.co.jp*

Observations of low energy nuclear reactions induced by D<sub>2</sub> gas permeation through Pd complexes (Pd/CaO/Pd) were presented at ICCF-9<sup>1</sup> and in a paper<sup>2</sup> published in the Japanese Journal of Applied Physics (JJAP). When Cs was added on the surface of a Pd complex, Pr emerged on the surface while Cs decreased after the Pd complex was subjected to D<sub>2</sub> gas permeation. When Sr was added to the surface, Mo emerged while the Sr decreased after D<sub>2</sub> gas permeation. The isotopic composition of the detected Mo was different from the natural abundance.

In this paper, recent progress of our research is described. The detected Pr was confirmed by various methods such as TOF-SIMS, XANES, X-ray Fluorescence Spectrometry and ICP-MS. Analysis of the depth profile of Pr indicated that a very thin surface region up to 100 angstroms was the active transmutation zone. Many experimental results showed that the quantity of Pr was proportional to the deuterium flux through Pd complex. The cross section of transmutation of Cs into Pr can be roughly estimated at 1 barn if we consider the deuterium flux as an ultra low energy deuteron beam.

### 1 Introduction

Anomalous elemental changes have been observed on the Pd complexes, which consist of a thin Pd layer, alternating CaO and Pd layers and bulk Pd, after subjecting the Pd complexes to D<sub>2</sub> gas permeation as we reported at ICCF-9<sup>1</sup> and in the paper<sup>2</sup> in the Japanese Journal of Applied Physics (JJAP).

In this paper, we describe recent progress. The following points have been improved or changed.

- 1) Pr was identified by XPS, TOF-SIMS, XANES, X-ray Fluorescence and ICP-MS.
- 2) A quantitative analysis of Pr has become possible using ICP-MS.
- 3) The correlation between deuterium flux and Pr was investigated.
- 4) Cs ion injection into Pd complexes (instead of the electrochemical method) was performed.
- 5) Depth profile and surface distribution of Cs and Pr was obtained by TOF-SIMS (Time of Flight Secondary Ion Mass Spectrometry).

Our experimental method can be characterized by the following two main features. The first is the permeation of D<sub>2</sub> gas through the Pd complex, as shown in Fig. 1(a). Permeation of deuterium is attained by exposing one side of the Pd complex to D<sub>2</sub> gas while maintaining the other side under vacuum conditions. On the D<sub>2</sub> gas side of the Pd complex, dissociative absorption causes the D<sub>2</sub> molecules to separate into D atoms, which diffuse through the metal toward the vacuum side, where they emerge from the metal, combine and are released as D<sub>2</sub> gas.

The second feature is the addition of an element that is specifically targeted to be transmuted. Our sample is a Pd complex composed of bulk Pd on the bottom, alternating CaO and Pd layers, and a Pd thin film on top. After fabricating a Pd complex, Cs or Sr is deposited on the surface of the top thin Pd layer, as shown in Fig. 1(b). This Cs or Sr is transmuted. In other words, with this composition, we can provide a deuterium flux through the Pd complex on which a target element is placed as a target to be transmuted. We perform elemental analyses of the given elements after D<sub>2</sub> gas permeation by exhausting the D<sub>2</sub> chamber (by making it into a vacuum chamber). Our experimental method is superior in that it clearly discriminates transmutation products from

contamination because we analyze the products by XPS (X-ray Photoelectron Spectroscopy) in vacuum, in situ during the experiment, without moving or the sample or opening the chamber.

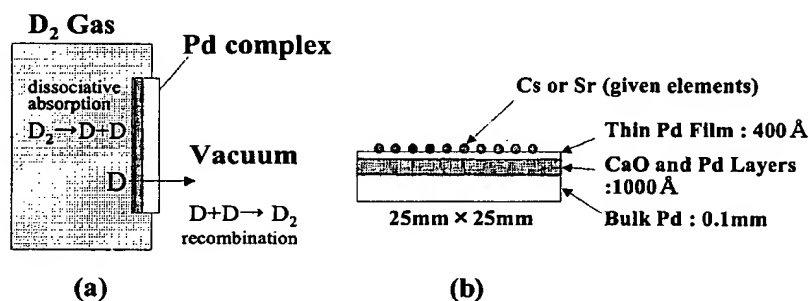


Figure 1. Schematic of the present method: (a) D<sub>2</sub> gas permeation of the Pd complex, (b) Structure of the Pd complex deposited with Cs or Sr

## 2 Experimental

The experimental method and setup are basically the same as before<sup>1,2</sup>. Therefore we shall omit a detailed description, and describe only the changed and improved aspects of the experiment.

Cs is now added to the surface by the ion injection method, in addition to the electrochemical method, for exact depth profile analysis.

Figure 2 shows the experimental apparatus. The D<sub>2</sub> gas flow rate was estimated by measuring the pressure of the chamber B. (Chamber B is evacuated, but the vacuum is gradually filled with the gas that permeates through the Pd complex.) The calibration curve for pressure versus the D<sub>2</sub> gas flow rate was obtained in advance by letting D<sub>2</sub> gas into the vacuum chamber through a precision gas flow meter.

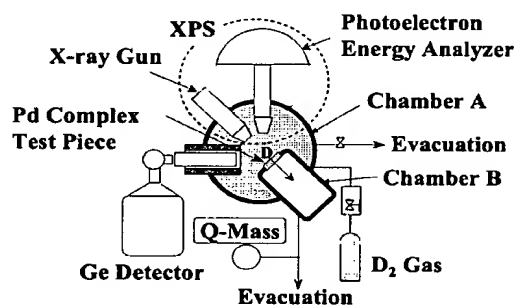


Figure 2. Experimental Setup.

### 3 Results and Discussion

Let us briefly describe the experimental results presented<sup>2</sup> at ICCF-9. A transmutation reaction converting Cs into Pr is shown in Fig. 3. Results for two runs are shown as examples. The number of Cs atoms decreased while the number of Pr atoms increased over time. No Pr was detected at the beginning of the experiments. At 120 h, the number of Pr atoms exceeded that of Cs atoms.

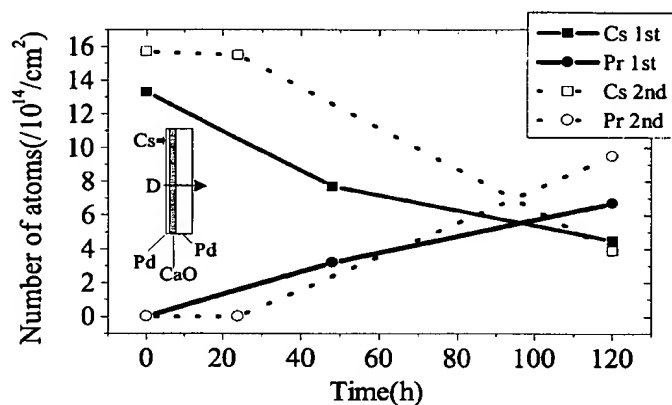


Figure 3. Time variation in the number of Cs and Pr atoms during D<sub>2</sub> gas permeation through Pd complex (Pd/CaO/Pd) deposited with Cs

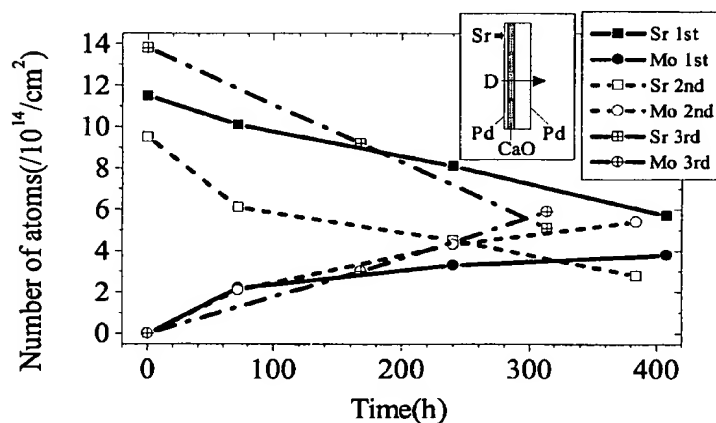


Figure 4. Time variation in number of Sr and Mo atoms induced by D<sub>2</sub> gas permeation through Pd complex (Pd/CaO/Pd) deposited with Sr

The experimental results for Pd complex test pieces with added Sr are shown in Fig. 4. We observed that Sr decreased while Mo increased over time. Experiments were performed three times and all data are plotted here. At the beginning of the experiments, no Mo atoms were detected. However, Mo atoms increased gradually while Sr decreased correspondingly. It should be noted that runs with Sr take longer to convert a given mass of Sr into Mo than it takes to convert that mass of Cs into Pr.

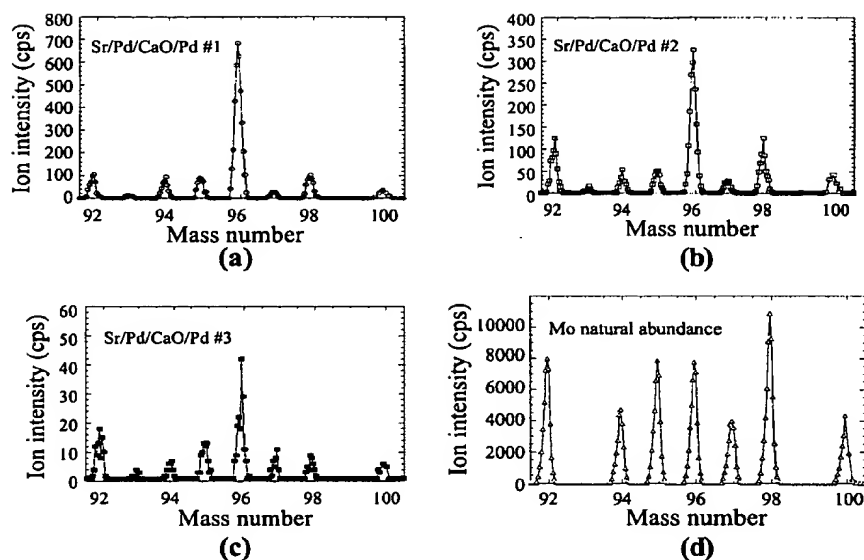


Figure 5. Anomalous isotopic composition of detected Mo: (a) Isotopic composition of detected Mo for run #1, (b) Isotopic composition of detected Mo for run #2, (c) Isotopic composition of detected Mo for run #3, (d) Natural abundance of Mo analyzed by SIMS.

Figures 5(a)–(c) show the results of SIMS analysis for the three samples. The intensities of mass number 96 were the largest for each sample, although the intensities were different. The SIMS mass spectrum for a Mo layer (400 Å thickness) that was deposited on a Pd disk is shown in Fig. 5(d). This spectrum reveals the natural abundance of Mo. Comparing these figures, we can easily recognize that the isotopic compositions of the detected Mo are different from the natural isotopic abundance of Mo.

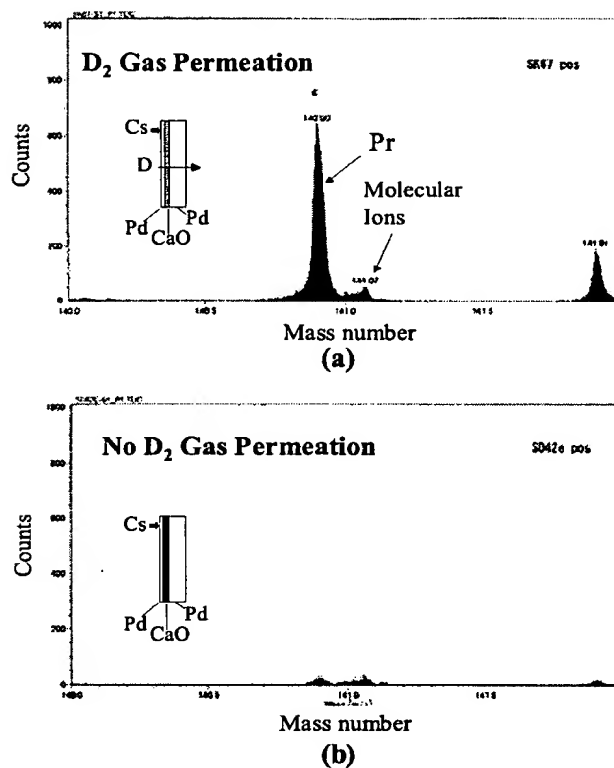


Figure 6. Identification of Pr by TOF-SIMS: (a) Mass number distribution of the sample after D<sub>2</sub> gas permeation, (b) Mass number distribution of the sample without D<sub>2</sub> gas permeation

Let us move on to new experimental results. Pr, the transmuted product from Cs, was confirmed by many element analysis methods. The first example is the identification of Pr by TOF-SIMS (Time of Flight Secondary Ion Mass Spectrometry) shown in Fig. 6. The TOF-SIMS device is a model TRIFTM II made by ULVAC-PHI. The upper figure shows the mass number distribution of the Pd complex (Pd/CaO/Pd) after D<sub>2</sub> gas permeation, and the lower figure is for the Pd complex without D<sub>2</sub> gas permeation. The TOF-SIMS can distinguish small mass difference so that Pr and molecular ions can be clearly separated, as shown in the upper figure. It is confirmed that Pr is detected only for the foreground sample.

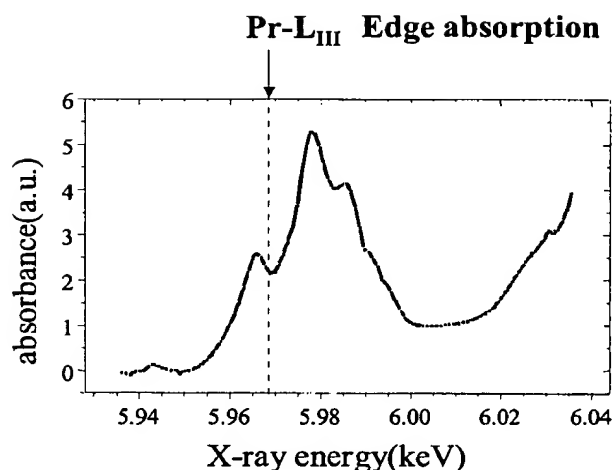


Figure 7. Identification of Pr by XANES (X-ray Absorption Near Edge Structure) .

Confirmation of Pr by XANES (X-ray Absorption Near Edge Structure) is shown in Fig. 7. This spectrum was obtained at the BL-9A Line at the High Energy Accelerator Research Organization (KEK), located in Tsukuba, Japan ([www.kek.jp](http://www.kek.jp)). A Pd complex sample after D<sub>2</sub> gas permeation, on which Pr was detected by XPS, was examined by XANES. Pr L<sub>III</sub> Edge absorption was clearly recognized in Fig. 7

Furthermore, Pd complex samples after D<sub>2</sub> gas permeation were examined by X-ray fluorescence spectrometry and ICP-MS (Inductively Coupled Plasma Mass Spectrometry). Although the X-ray fluorescence spectrometry is a bulk analysis method, Pr was detected using strong SOR X-rays. The sensitivity of ICP-MS is so high that quantitative analysis of Pr is performed for all the experiments.

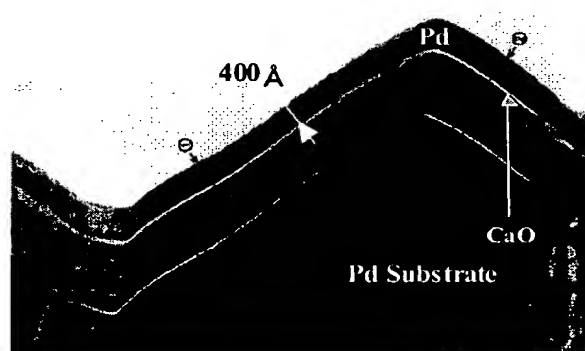


Figure 8. Cross section of Pd complex (Pd/CaO/Pd) observed by TEM (Transmission Electron Microscopy)

Figure 8 shows the cross sectional view of the Pd complex (Pd/CaO/Pd). This image was taken by TEM (Transmission Electron Microprobe). During the process of Pd complex fabrication, the Pd substrate is etched with aqua regia<sup>1,2</sup>. The wave-like shape of the Pd substrate is formed by the etching process. On the Pd

substrate, Pd and CaO complex layer are formed by Ar ion beam sputtering. The white lines correspond to CaO and the black parts to Pd. The 400-angstrom Pd thin film is located on the Pd and CaO complex layer.

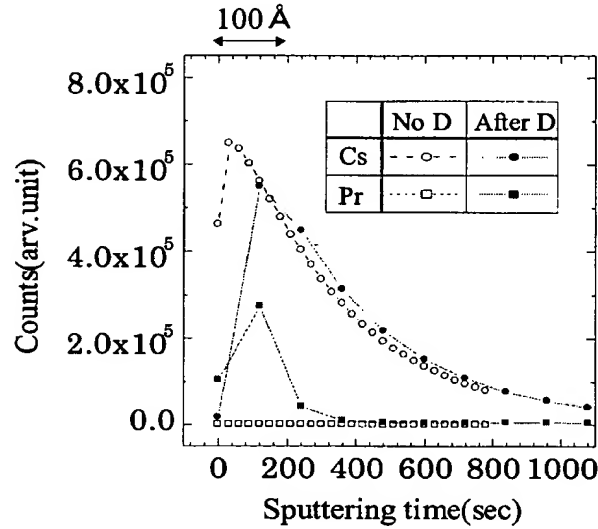


Figure 9. Depth Profiles of Cs and Pr for a Pd complex (Pd /CaO/Pd) sample after D<sub>2</sub> gas permeation and a Pd complex (Pd /CaO/Pd) sample without D<sub>2</sub> gas permeation

Depth profiles of Cs and Pr were plotted in Fig. 9. Two Pd complex samples were prepared and Cs was injected into them by the ion implantation method. Acceleration voltage and Cs fluence for the ion implantation were the same for the two samples, 18keV and  $10^{15}$  ions/cm<sup>2</sup>, respectively. The depth profiles were estimated by TOF-SIMS analysis. Physical Electronics TRIFT II was applied for the analysis and the condition of Ga<sup>+</sup> ion was 15keV-600pA. The relation between the sputtering time and the real depth was estimated in advance using a Pd thin film on Si substrate; thickness of the Pd thin film is known. This measurement shows that a 200 sec sputtering time corresponds to 100 angstroms.

Cs and Pr depth profiles for the Pd complex without permeation show normal results in Fig. 9. Cs decreases continuously from the surface and there is no Pr in the sample.

On the other hand, Cs and Pr depth profiles for the Pd complex after D<sub>2</sub> gas permeation exhibit interesting results. Cs depth profiles for the foreground and background samples agree in the deep area. However, Cs decreases near the surface after D<sub>2</sub> gas permeation. We can see that there is Pr, which is the same order as given Cs, in the near surface area. This experimental fact suggests that Cs transmutation reaction into Pr occurs in the near surface region up to 100 angstrom. This transmutation active zone might be correlated with the D/Pd ratio. Further investigation of the surface region is important. Figure 9 also shows that Cs atoms do not diffuse and migrate with D<sub>2</sub> gas permeation under our experimental conditions. Therefore it is very difficult to imagine that the detected Pr was a concentrated impurity, and not a transmutation product.

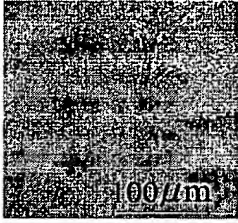

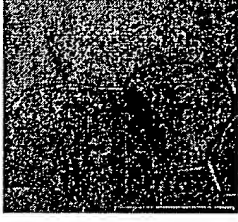

	Cs	Pr
before	 A grayscale micrograph showing the surface distribution of Cs before D2 gas permeation. The image displays a granular texture with some darker regions. A scale bar at the bottom right indicates 100 μm.	 A grayscale micrograph showing the surface distribution of Pr before D2 gas permeation. The image is mostly black, indicating a very low or uniform concentration of Pr.
after	 A grayscale micrograph showing the surface distribution of Cs after D2 gas permeation. The image shows a more pronounced granular texture compared to the 'before' state.	 A grayscale micrograph showing the surface distribution of Pr after D2 gas permeation. The image shows a more uniform distribution of Pr across the surface compared to the 'before' state.

Figure 10. Surface Distributions of Cs and Pr for a Pd complex (Pd /CaO/Pd) sample after D<sub>2</sub> gas permeation and a Pd complex (Pd /CaO/Pd) sample without (before) D<sub>2</sub> gas permeation

Figure 10 shows surface distributions of Cs and Pr for the two samples discussed above. Space resolving power is 1 micron. Grain boundaries can be seen in each image. These images show that the surface distribution of Pr basically seems to be uniform and has no correlation with the grain boundaries.



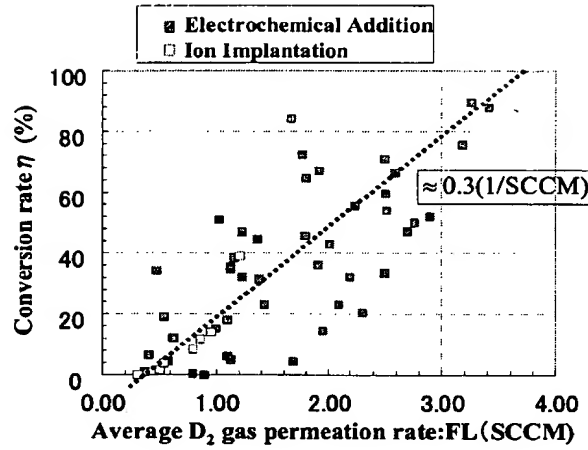


Figure 11. Correlation between D<sub>2</sub> permeation rate and conversion rate.

Using ICP-MS analysis, a quantitative estimate of the mass of Pr has been performed. The correlation between D<sub>2</sub> gas permeation rate and conversion rate is shown in Fig. 11.

The conversion rate is defined as

$$\eta = \frac{N_{Pr}}{N_{Cs}} \times 100\% = \frac{N_{Pr}}{N'_{Cs} + N_{Pr}} \times 100\%,$$

$\eta$  : conversion rate(%),  $N_{Pr}$  : detected Pr(ng),  $N_{Cs}$  : given Cs(ng),

$N'_{Cs}$  : detected Cs after an experiment(ng).

(1)

Since ICP-MS analysis is a destructive analysis method, we cannot measure the starting mass of Cs directly. Assuming that a Cs atom is transmuted into a Pr atom, the sum of the detected Pr and the detected Cs after permeation should be equal to the starting Cs.

Figure 11 suggests that the conversion rate defined as above is proportional to the average D<sub>2</sub> gas permeation rate. Experimental results for both the electrochemical addition and the ion implantation of Cs are plotted in the figure. It seems that they have linear correlation for the both cases.

Let us consider on the situation that D beam irradiates the Pd complex with Cs. The reaction rate is expressed as the following equation.

$$R = \sigma \cdot N_{Cs} \cdot \phi$$

$R$  : reaction rate(event/cm<sup>3</sup>/sec),  $\sigma$  : cross section(cm<sup>2</sup>),  $N_{Cs}$  : number of Cs(1/cm<sup>3</sup>),

$\phi$  : deuteron beam flux (1/cm<sup>2</sup>/sec).

(2)

If we regard D<sub>2</sub> gas permeation as a kind of deuteron beam, the following relation is obtained by equation (2).

$$\eta = \int_t (R/N_{Cs}) dt = \int_t (\sigma \cdot \phi) dt = \sigma \int_t \phi dt \approx \sigma \cdot f \cdot FL \cdot T_{exp} / S \quad (3)$$

FL : flow rate(sccm),  $T_{exp}$  : reaction time(sec), S : permeation surface area( $cm^2$ ).

$$\therefore \eta \propto FL$$

This equation agrees with the experimental results shown in Fig. 11. Therefore we can roughly estimate the cross section using the obtained experimental results. If we input experimental parameters into equation (3), we obtain

$$\begin{aligned} \eta &= \sigma \cdot f \cdot FL \cdot T_{exp} / S \\ &= \sigma \cdot \frac{2 \times 6 \times 10^{23}}{22.4 \times 10^3 \times 60} \cdot FL \cdot 100 \times 3600 / 1.0 \\ &= \sigma [cm^2] \cdot FL [sccm] \cdot 3 \times 10^{23} [1/cm^2 / sccm] \end{aligned} \quad (4)$$

The experimental results show the gradient between FL and conversion rate is about 0.3(1/sccm). (The term sccm means standard cubic centimeter per minute.) Therefore the following result is obtained.

$$0.3 \approx \sigma \cdot 3 \times 10^{23} \quad \therefore \sigma \approx 1 \times 10^{-24} [cm^2] = 1[barn]$$

This cross section seems to be extremely large if we take it into consideration that the transmutation reaction belongs to multi-body reactions. And we should notice that we regard the deuterium permeation velocity as deuteron velocity, and the deuteron flux is estimated relatively low, leading to very large cross section. If the deuteron behavior on the microscopic level in the Pd thin film could be clarified, a more precise physical model would be developed. In any case, on the macroscopic level deuterium permeation through Pd complex can be regarded similar to an ultra low deuteron beam, and the cross section of transmutation of Cs into Pr is estimated at 1 barn according to our experimental results.

We would like briefly touch on a few points, starting with the problem of discriminating contamination and transmutation products. Since the detected material, Pr, is a rare earth element, it is difficult to imagine that Pr accumulated on the Pd complex test samples by any ordinary process. As mentioned in Fig. 9, Cs atoms do not diffuse and migrate by  $D_2$  gas permeation. Therefore it can be postulated that Pr atoms also do not migrate. The purity of our  $D_2$  gas is over 99.6% and the most of the impurity in it is  $H_2$ . Other impurities detected by a mass spectrometer are  $N_2$ ,  $D_2O$ ,  $O_2$ ,  $CO_2$ ,  $CO$  and hydrocarbons; they are all under 10ppm. We analyzed Pd complex test pieces deposited with Cs by ICP-MS mass spectrometry and confirmed that Pr in the test samples was below the detection limit (0.1ng). On the other hand, the detected Pr ranges from 1ng to 100ng. The amount of the detected Pr exceeds the maximum possible contamination of Pr. Therefore we conclude the detected Pr was transmuted from Cs.

Our next point is that the isotope ratio of the synthesized elements is anomalous. The isotopic anomaly of the Mo is particularly strong evidence that this Mo was produced by some nuclear processes. Some might speculate that the anomalous isotope ratios were caused by Mo contamination undergoing some sort of isotopic separation process, leaving only  $^{96}Mo$  to be detected. (See Fig. 5). However, such efficient isotope separation would not be possible.

We noticed that a certain rule exists between starting and produced elements<sup>1,2</sup>. The increase in mass number is 8, and the increase in atomic number is 4 in the case of Cs and Sr. It appears that 4d addition reactions occur. We also observed 2d and 6d addition transmutation reactions<sup>3</sup>.

At present, we do not have a complete theory that can explain the experimental results without a few assumptions. The EQPET model<sup>4,5</sup> proposed by Prof. A. Takahashi can basically explain our experimental results, by assuming that a short lived, quasi-particle electron pair like Cooper-pair can be generated. The observed transmutation processes must belong to a new category of nuclear reactions in condensed matter. Therefore much more theoretical investigation is necessary.

#### 4 Concluding Remarks

Nuclear transmutation of Cs into Pr and Sr into Mo can be observed during D<sub>2</sub> gas permeation through Pd Complexes. Pr was identified by various methods such as XPS, TOF-SIMS, XANES, X-ray fluorescence spectrometry and ICP-MS. A very thin surface region up to 100 angstroms was the active transmutation area, as determined by the analysis of depth profile of Pr. The quantity of Pr was proportional to deuterium flux through the Pd complex. The cross section of transmutation of Cs into Pr can be roughly estimated at 1 barn if we regard the deuterium flux as an ultra low energy deuteron beam.

Some replication experiments producing transmutation reactions of Cs into Pr or Sr into Mo were planning or presented for the ICCF10 conference<sup>6,7</sup>. Positive results were obtained not only in a gaseous environment<sup>6</sup> presented by Prof. A. Takahashi *et al.*, but also in an electrochemical environment<sup>7</sup> performed Dr. F. Celani's team.

#### Acknowledgments

The authors would like to acknowledge Prof. A. Takahashi, Dr. F. Celani, Dr. I. Tanihata, Dr. T. Ishikawa, Dr. Y. Terada, Dr. K.S. Grabowski, Dr. G.K. Hubler, Prof. T. Okano, Dr. K. Fukutani, Prof. S. Tanaka, Prof. K. Okuno and Prof. J. Kasagi for their valuable discussions.

#### References

1. Y. Iwamura, M. Sakano and T. Itoh, *Elemental Analysis of Pd Complexes: Effects of D<sub>2</sub> gas permeation*. Jpn. J. Appl. Phys. **41** (2002), pp. 4642-4648.
2. Y. Iwamura, T. Itoh, M. Sakano and S. Sakai, *Observation of Low Energy Nuclear Reactions induced by D<sub>2</sub> gas permeation through Pd Complexes*. Proc. of ICCF9 19-24 May 2002, Beijing (China); pp.141-146.
3. Y. Iwamura, T. Itoh and M. Sakano, *Nuclear Products and Their Time Dependence Induced by Continuous Diffusion of Deuterium through Multi-layer Palladium Containing Low Work Function*. Proc. of ICCF8, 21-26 May 2000 Lerici (Italy), SIF Conf. Proc. Vol.70, pp.141-146.
4. A. Takahashi, *Tetrahedral and octahedral resonance fusion under transient condensation of deuterons at lattice focal points*. Proc. of ICCF9 19-24 May 2002, Beijing (China); pp.343-348.
5. A. Takahashi, *Mechanism of Deuteron Cluster Fusion by EQPET model*. Proc. of ICCF10, in press.
6. T. Higashiyama *et al.*, *Replication of MHI transmutation experiment by D<sub>2</sub> gas permeation through Pd complex*. Proc. of ICCF10, in press.
7. F. Celani *et al.*, *Thermal and Isotopic Anomalies when Pd Cathodes are Electrolysed in Electrolytes Containing Th-Hg Salts Dissolved at Micromolar Concentration in C<sub>2</sub>H<sub>5</sub>OD/D<sub>2</sub>O Mixtures*. Proc. of ICCF10, in press.

Higashiyama, T., et al. *Replication Of MHI Transmutation Experiment By D<sub>2</sub> Gas Permeation Through Pd Complex*. in *Tenth International Conference on Cold Fusion*. 2003. Cambridge, MA: LENR-CANR.org. This paper was presented at the 10th International Conference on Cold Fusion. It may be different from the version published by World Scientific, Inc (2003) in the official Proceedings of the conference.

## Replication Of MHI Transmutation Experiment By D<sub>2</sub> Gas Permeation Through Pd Complex

Taichi Higashiyama\*, Mitsuru Sakano\*\*,  
Hiroyuki Miyamaru\* and Akito Takahashi\*

\*Osaka University, Yamadaoka 2-1, Suita, Osaka, 565-0871 Japan

\*\*Mitsubishi Heavy Industry, Sachiura 1-8-1, Kanazawa-ku, Yokohama, 236-8515 Japan  
[higashiyama@stu.nucl.eng.osaka-u.ac.jp](mailto:higashiyama@stu.nucl.eng.osaka-u.ac.jp)

Unusual nuclear transmutation reactions have been reported by Mitsubishi Heavy Industries (MHI). In their experiment, D<sub>2</sub> gas permeates through a Pd complexes, which consists of a thin Pd layer, alternating CaO and Pd layers and bulk Pd.<sup>1</sup> When they used sample Pd complexes with additional Cs on the surface, Pr emerged on the surface while Cs decreased after the sample was subjected to D<sub>2</sub> gas permeation at 343 K and 1 atm for about one week. The elemental analysis was performed by X-ray photoelectron spectroscopy (XPS). This phenomenon was reproduced qualitatively in the present replication experiment.

We performed D-permeation experiments similar to the MHI's experiment<sup>1</sup> three times, and we confirmed the production of Pr. Pd complex samples were provided to us by MHI. The surface was electrolytically cleaned to remove hydrocarbons before depositing Cs. D<sub>2</sub> gas was permeated through the Pd complexes at 343 K and 1 atm for about 5 days. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) was performed to analyze the existence of the elements (Cs and Pr) and the mass distribution. The results showed the existence of Pr. And we also confirmed the existence of Pr by using fast Neutron Activation Analysis (NAA) in Fusion Neutronics Source (FNS) of Japan Atomic Energy Research Institute (JAERI).

As a result, we confirmed that the nuclear transmutation reaction, from <sup>133</sup>Cs to <sup>141</sup>Pr, was occurred. This transmutation suggests that the mass numbers and atomic numbers increase 8 and 4, respectively. The model of multi-body resonance fusion of deuterons proposed by A. Takahashi<sup>2</sup> can explain this mass-8-and-charge-4 increased transmutation.

### 1 Introduction

We received a report from Iwamura's group that unusual nuclear reactions were caused by permeating D<sub>2</sub> gas through Pd complexes with added Cs or Sr on the surface<sup>1</sup>. The condition was that D<sub>2</sub> gas flow was over 1 sccm, the temperature was 343 K, D<sub>2</sub> gas pressure was 1 atm and the flow time was 1 week. (Note: the unit "sccm" is a flow rate of one cc per a minute in standard conditions.)

In contrast, permeating H<sub>2</sub> gas instead of D<sub>2</sub> gas showed no decreasing of Cs and no generation of Pr. The elemental analysis was done by XPS.

Following the MHI procedure exactly, we performed D<sub>2</sub> gas permeation experiments through Pd complexes and analyzed the production of Pr. The following is the first report of this replication experiment.

### 2 Experiments

#### 2.1 Samples

Pd complex samples were composed of a Pd thin film, alternating CaO and Pd layers and bulk substratum Pd as shown in Fig. 1. The surface of the substratum Pd plate was first covered by alternative layers of CaO and Pd

(1000Å). Then a 400-Å-thick Pd layer was sputtered on the surface of the alternative CaO and Pd layers. These processes were performed by Ar ion beam sputtering. After forming a Pd complex, Cs was deposited on the surface of the thin Pd layer. Cs atom was deposited by applying a weak electric field to 1 mM CsNO<sub>3</sub> solution. A 1 V negative voltage was applied to the Pd complex for 10 seconds.

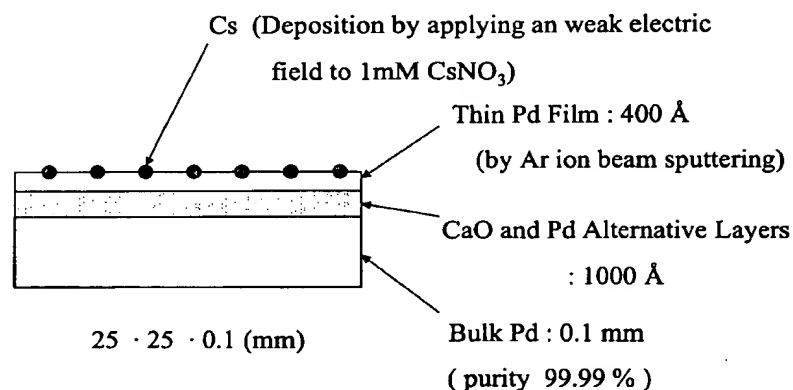


Figure 1. Sample fabricated by MHI

To remove surface hydrocarbon contamination from on the sample, we performed following pre-cleaning operation before putting Cs on the sample. The sample and a Pt plate were applied with an electric field to 1mM HNO<sub>3</sub>. Then the bubble was formed on the surface of the Cs deposit side. And Teflon was used to set up the sample and the Pt plate.

## 2.2 Experimental equipments

Chamber A was filled with 1 atm D<sub>2</sub> gas, and Chamber B was evacuated by a turbo-molecular pump (TMP) as shown in Fig. 2, so that the front surface of a Pd complex sample was in 1 atm D<sub>2</sub> gas and the back surface was on vacuum. D<sub>2</sub> gas was supplied at 1 atm on the Cs side of the sample so that deuterium atoms permeate to the backside of the sample in Chamber B. The sample was normally heated to 70 degree Celsius during these experiments.

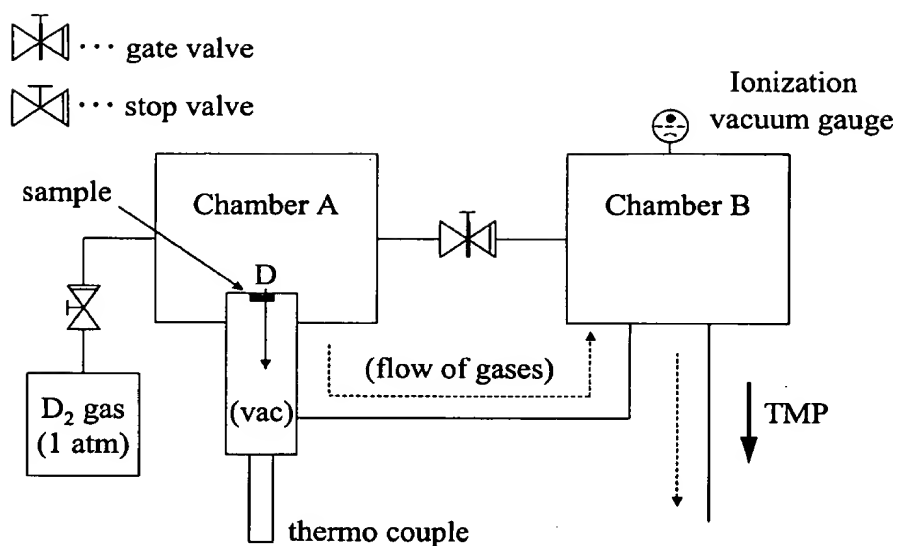


Figure 2. Experimental equipments

### 2.3 D<sub>2</sub> gas permeation experiments

#### 2.3.1 The first run

Using the experimental system shown in Fig. 2, we have performed three experimental runs of D<sub>2</sub> gas permeation through Pd complexes. We changed the temperature stepwise to 70, 60 and 80 degree Celsius and permeated D<sub>2</sub> gas for about 120 h in the first run.

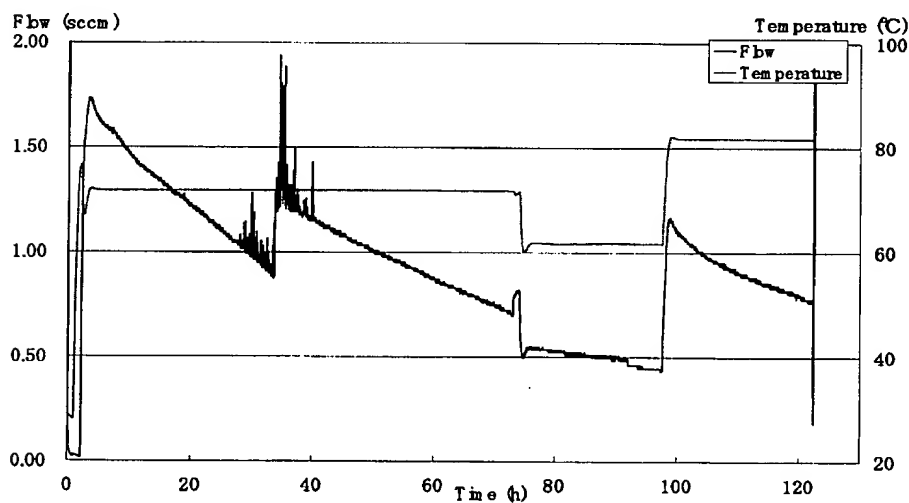


Figure 3. Relation between deuterium flow rate and temperature for the first run

We show the result in Fig. 3. The flow rate exceeded 1.5 sccm initially, decreased slowly, and then fell below 1.0 sccm, which is considered the critical value for transmutation, according to the MHI group. The flow rate increased as the temperature was stepped up, and exceeded 1 sccm again, but it soon declined again. We observed unnatural fluctuations in the flow rate between 20 h and 40 h. We assume that electrical miscontact of the ionization vacuum gauge caused this.

### 2.3.2 The second run

In the second run, the flow rate was below 1 sccm from the beginning, so we raised the temperature from 70 to 80 degrees Celsius and permeated  $D_2$  gas for about 200 h in this experiment, as shown in Fig. 4. But the flow was still below 1 sccm. We consider the likely reason is that moisture from the atmosphere came inside the chamber and formed on the sample surface, which prevented  $D_2$  gas from permeating through the sample efficiently. The data gap near 30 h occurred when we stopped the vacuum equipment because of a shortage of  $N_2$  gas.

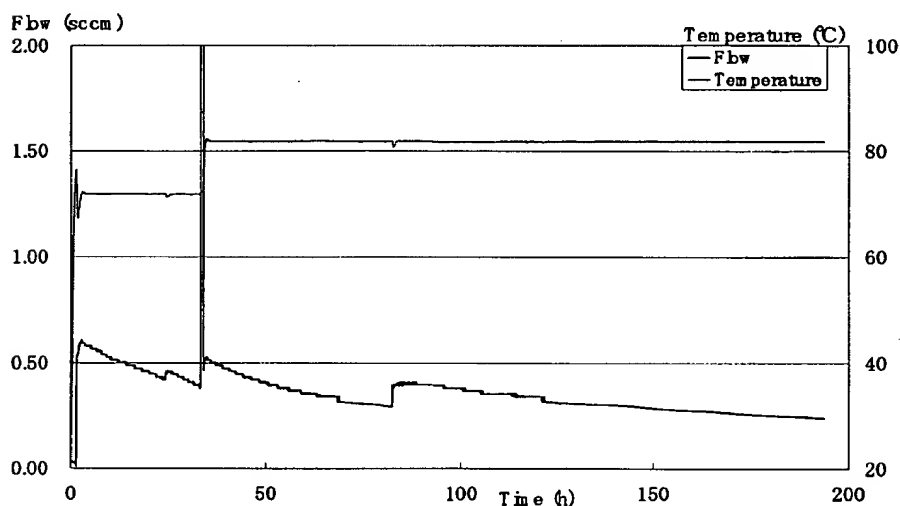


Figure 4. Relation between deuterium flow rate and temperature for the second run

### 2.3.3 The third run

We kept the temperature constant at 70 degree Celsius and permeated  $D_2$  gas for about 120 h in the third run. To keep the sample clean, and the chamber free of moisture from the atmosphere, we first baked out the chamber, and then  $N_2$  gas was fed and evacuated before setting up the sample in the chamber. As the result, flow rate was improved to over 2 sccm at first. But because we did not have enough remaining  $D_2$  gas, the pressure in the chamber filled with  $D_2$  gas could not be maintained at 1 atm and the flow rate decreased gradually. The detailed flow rate data before 40 h was lost because of a problem in the data acquisition system, so the curve is approximated with data from 0 h and 20 h taken manually from the ionization vacuum gauge, as shown in Fig. 5.

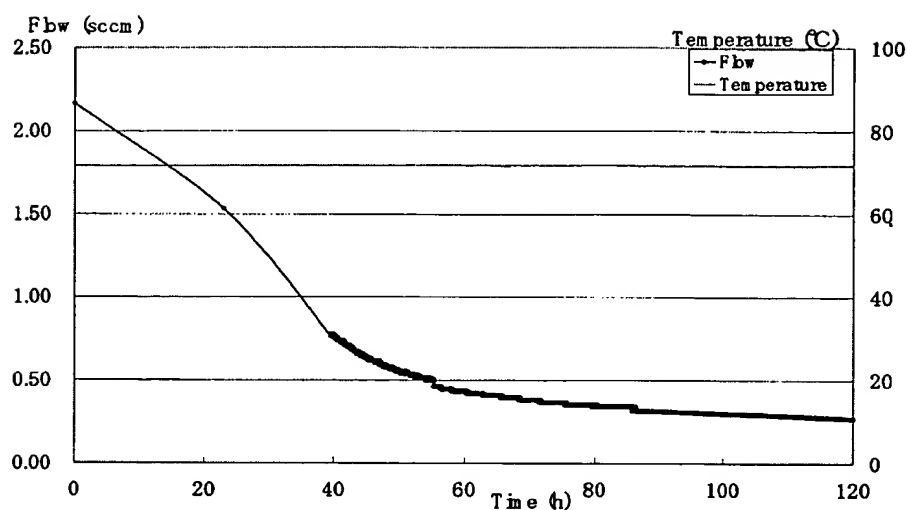


Figure 5. Relation between deuterium flow rate and temperature for the third run

### 3 Results of Element Analysis

#### 3.1 Analysis by ICP-MS

The elements on the surface of the sample were analyzed by ICP-MS in the MHI laboratory. The result revealed the production of Pr atoms, although amounts were different with each run, as shown in Table 1. But a correlation exists between the amounts of Pr produced and the maximum flow rates.

Table 1. Results analyzed by ICP-MS in the MHI

	Pr (ng)	Cs (ng)	max flow(sccm)	average (sccm)	minimum(sccm)
1st run	18	180	1.73	0.93	0.43
2nd run	51	141	0.61	0.35	0.29
3rd run	36	330	2.17	0.76	0.27

#### 3.2 Analysis by NAA

We also analyzed the Pd complex sample by Neutron Activation Analysis (NAA) using 14 MeV neutrons in Fusion Neutronics Source (FNS) of Japan Atomic Energy Research Institute (JAERI), for the sample that was permeated D<sub>2</sub> gas in MHI and analyzed by XPS in MHI<sup>1</sup>.

Figure 6 shows the result of Ge gamma-ray spectra by 14 MeV fast neutron irradiation in the FNS. In the first measurement, the sample was irradiated for 11 days, cooled for 25 days and measured for 7 days. And at the second measurement, taken from the same sample after the first measurement, the sample was cooled for 8 days



and measured for more 9 days. We increased the HPGe detector efficiency by setting the sample very close to the detector surface for the second measurement, to improve counting statistics.

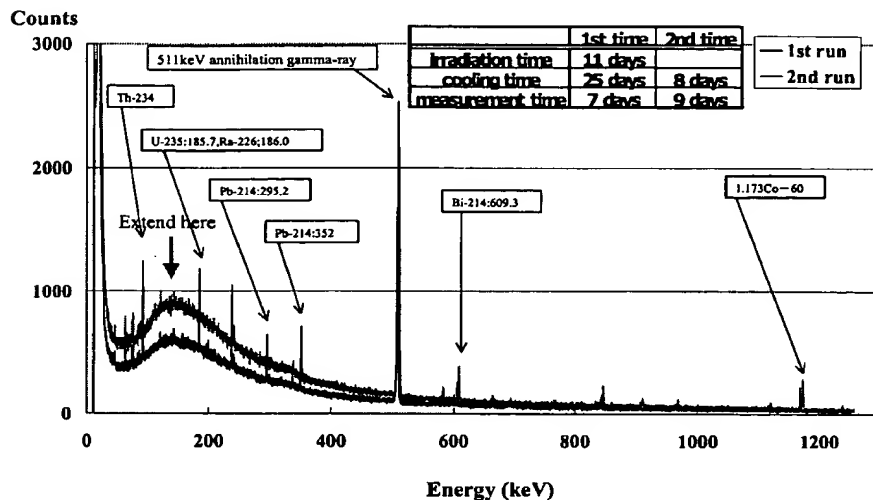


Figure 6. Ge gamma-ray spectra by NAA of 14 MeV neutron irradiation

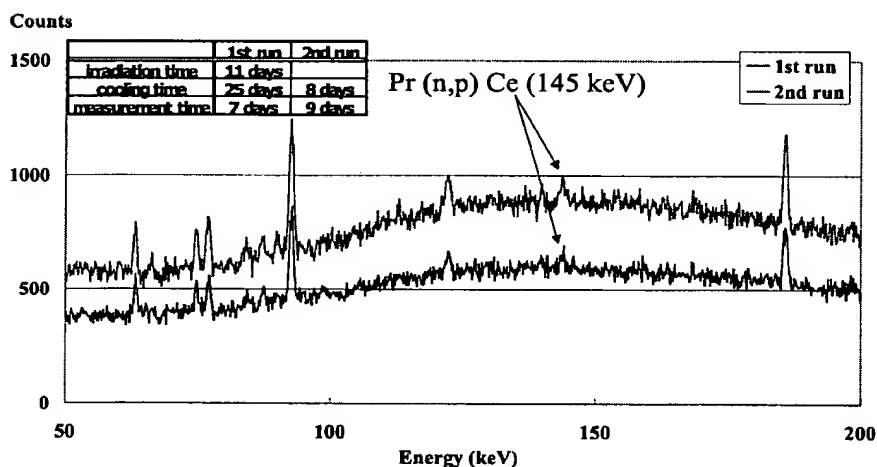


Figure 7. Expanded spectra for detail analysis by NAA of 14 MeV neutron

Figure 7 shows the region near 145 keV which indicates the existence of Pr by the gamma-ray peak at 145 keV of  $^{141}\text{Pr}(n,p)^{141}\text{Ce}$  reaction. For the second measurement, we increased Ge detector efficiency, so that we compared two (first and second measurement) spectra after the normalization with 511 keV peaks of

background gamma-rays. We could see a decay of 145 keV peaks of  $^{141}\text{Ce}$  from the first measurement to the second measurement, with half-life of about 50 days with large (more than 50 %) error bar obtained by only two data points (32.5 days is the exact half-life). This 145 keV peak with decay convinced us of the existence of  $^{141}\text{Pr}$  in the sample. However the counting statistics for peaks of  $^{141}\text{Ce}$  were not high enough to obtain the exact half-life. We are planning another irradiation to improve the statistics.

#### 4 Discussion and conclusion

After the three runs of experiments permeating  $\text{D}_2$  gas through in our laboratory, samples were analyzed by ICP-MS in MHI and we could confirm existence of Pr from three runs, as shown in Table 1.

And we also confirmed the existence of Pr by 14 MeV NAA in FNS as shown in Fig. 7, for the sample which  $\text{D}_2$  gas permeation was done by MHI.

In order to explain this phenomenon that Cs changes to Pr, a "theory of 8D nuclear fusion" is considered<sup>2</sup>. This model holds that two high energy Be-8 nuclei ( $\text{Be}^*$ ) are produced by octahedral resonance fusion of eight deuterons in the Pd lattice, and these Be particles may be absorbed by Cs (or Sr) to cause transmutation with mass number 8 and atomic number 4 increased reaction, as shown Fig. 8. Also, these Be-8 nuclei have very short lifetime  $6.7 \cdot 10^{-17}$  s, and they decay to two stable He atoms as soon as they are formed, but we have enough collisions with Cs (or Sr) nuclei within range.

Reaction formula is as follows:

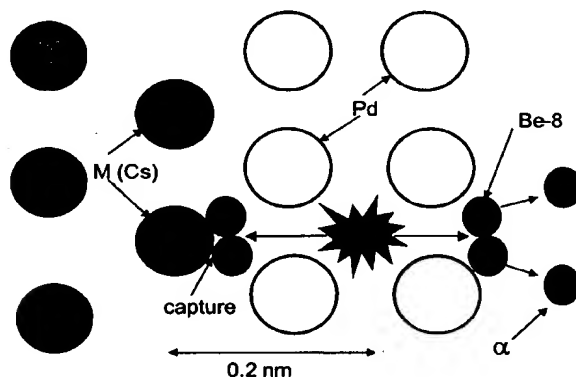
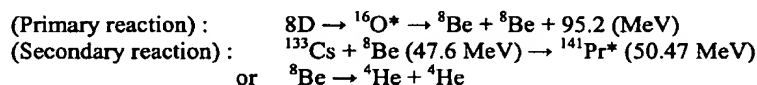


Figure 8. Model of 8D nuclear fusion

If nuclear transmutation is induced by the secondary reactions of 8D fusion,  ${}^4\text{He}$  gas must be produced. So we are planning an experiment to confirm  ${}^4\text{He}$  gas production by using a QMAS system.

#### Reference

1. Y. Iwamura et al., Proc. ICCF 9, May 19-23, 2002, Beijing, China, pp.141-146.
2. A. Takahashi, Proc. ICCF9, May 19-23, 2002, Beijing, China, pp.343-348.